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Performance Dependence of Organic Solar Cells as a Function of Active Layer Thickness, Luminous Intensity and Temperature of Thermal Annealing

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Photovoltaic cells are optoelectronic devices for converting light energy into electrical energy. This work studies the effect of the active layer thickness on the performances of the inverted organic solar cells. The structures of the devices manufactured are Glass/ITO/ZnO NPS/P3HT:PCBM/PEDOT(FO10)/Ag. The active layer based on a blend of poly (3-hexylthiophene) and fullerene with a ratio of 1:0.8 is spin-coating at different speeds in order to obtain different thicknesses. ZnO NPS (30 nm) were used to improve electron transport between ITO and the active layer. PEDOT (F010) (40 nm) were used as a hole transport layer. The commercial ITO used in this study has a sheet resistance of 7 Ω/□ for a thickness of 110 nm. The fabrication and the characterization of the cells as well as the optical properties of materials used and the photovoltaic parameters of the solar cells are reported in the paper. A direct relation between the active layer thickness and efficiency of the inverted organic solar cells is proved. An efficiency of 4.36 % is obtained for 200 nm of P3HT:PCBM. The effect of light intensity and the thermal annealing on the performances of photovoltaic cells are discussed. The characterizations of the cells show that the luminous intensity influences the electrical and optical parameters of organic solar cells.

Keywords: Inverted organic cells, Absorption, IPCE, Thickness, Light intensity, Thermal annealing.

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1. INTRODUCTION

Polymer solar cells (PSCs) have attracted much attention in recent years because of their advantages of simple device structure, low cost, light weight and flexibility [1]. The active layer of the PSCs is composed of a blend of a conjugated polymer donor and a fullerene derivative acceptor. P3HT and PCBM are the most representative donor and acceptor materials, respectively. Promising routes for increasing efficiencies and duration times based on the P3HT:PCBM blend have been envisaged [2]. P3HT exhibits a relatively high hole mobility (10^{-4}-10^{-3} $cm^2V^{-1}S^{-1}$) [3]. The active layer morphology related to blend preparation and annealing, is one of the most important factors affecting solar cells efficiency [4-6]. The thickness of the active layer coatings after spin-coating onto substrate is determined by the following parameters: spin speed, acceleration, spin time and substrate size.

Major improvements in terms of stability have been achieved by developing inverted OPV devices (IOSCs), where ITO is used as the cathode and a high work function Ag as the anode. In regular structure, holes and electrons are injected into transparent indium-tin oxide (ITO) and counter electrode (Al), respectively. In contrast, in an inverted device structure, electrons are injected into the ITO while holes are collected by the top electrode, which can be a less air sensitive, high work-function metal, such as silver or gold.

Transparent conducting oxides (TCOs) are widely used as electrodes in a large variety of optoelectronic devices due to their unique combination of optical and electrical properties (high transparency in the visible range and high electrical conductivity). Nowadays, indium tin oxide (ITO) is the most popular commercial TCO, having superior stability and conductivity. To improve the charge collection, functional layers are inserted to modify the interfaces of the active layers and electrodes.

The interface between the anode and the active layer should be significantly improved by a buffer layer such as PEDOT:PSS, V_2O_5, WO_3 and MoO_3 [14, 15]. PEDOT:PSS improves the performance of OSCs. PEDOT (F010) is particularly suitable in inverted cells on top of photo-active layers, such as P3HT:PCBM providing excellent surface wettability properties. The Clevios HTL is beneficial because: it forms ohmic contacts with TCOs and metal, it has a work function for efficient hole collection of about 5.0 eV, it reduces the probability of electrical shorts by smoothing defects on the surface of the substrate or sample and it is conductive and highly transparent.

To improve the electron transport between the ITO cathode and the active layer, an electron transport layer (ZnO NPs, TiOx, Ta_2O_5) is inserted.

In this paper, we present the obtained results related to the optical properties of P3HT:PCBM blends and the buffer layers used as ETL (ZnO NPs) or HTL (PEDOT (F010). We have examined the thickness-dependent device characteristics of BHJ-IOSCs with a P3HT:PCBM layer thickness of 190, 200 and 230 nm. The electrical characterizations of the cells were performed in an ambient atmosphere.

2. MATERIALS AND METHODS

The inverted bulk heterojunction organic solar cells BHJ-IOSCs are based on a blend of P3HT and PCBM. The glass substrate covered by transparent electrode of ITO with a sheet resistance of 7 Ω/square was ultrason-

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ically cleaned in acetone, ethanol and isopropanol for 10 min, respectively, and then treated by UV ozone for 15 min. The nanoparticles of zinc oxide ZnO NP used as electron transporting layer were spin-coated onto clean glass/ITO substrates (12 × 12 mm^2) using a speed of 2400 rpm for 30 s to obtain a layer of 30 nm, followed by annealing at 130 °C for 15 min to remove the solvent. The organic active layer P3HT:PCBM was coated on the glass/ITO/ZNO NPs substrate in the glove box under nitrogen atmosphere.

A blended solution containing 60 mg of poly (3-hexylthiophene) (P3HT, Reike Metals) and 48 mg of 1-(3-methoxycarbonyl)-propyl-1-phynyl-(6.6)C61 (PCBM, American Dye source) in 2 ml of 1.2-dichlorobenzene was prepared for 24 h before being used. After spin-coating the P3HT:PCBM active layer at various speeds in order to obtain different thicknesses, a solvent-annealing treatment was performed by keeping the active film inside a covered Petri dish for 120 min. Then a layer of PEDOT:PSS (Clevios F010) used as a hole transporting layer was spin-coated at 5500 rpm (40 nm). To complete the device, 120 nm-thick silver (Ag) anodes were deposited by thermal evaporation at a pressure of 10^{-6} mbar using a shadow mask of 0.18 cm.

The finished device has the structure as follows: glass/ITO/ZNO NPs/P3HT:PCBM (*x* nm)/PEDOT(F010)/Ag. The cells are annealed at 110°C for 30 min in the glove box before characterization. The thermal annealing process enhanced the crystallinity of P3HT due to the diffusion of PCBM molecules into aggregates and the P3HT crystallized in the PCBM free region [11]. For P3HT:PCBM annealing temperature affects the carrier mobility. Generally, electrons have higher mobility than the holes. The electron mobility of P3HT:PCBM after annealing at 120 °C is 3.10^{-7} $m^2V^{-1}S^{-1}$ [12].

The photocurrent density-voltage (*J-V*) curves were measured outside the glove box using a Keithley 2400 under simulated AM1.5 solar irradiance at 100 mW/cm^2. Solar radiation was calibrated using a standard silicon solar cell and a miss-factor of 0.8 (calculated using IPCE spectrum). The IPCE of the solar cells was taken under illumination from TS-428 Acton 250 W tungsten halogen lamp supplied by a JQE 25-10M Kepco voltage unit and monochromated by an Acton SpectraPro 2150i at 80 Hz chopping frequency. The photocurrent was detected by a Stanford Research SR530 lock-in amplifier and compared to the signal obtained under the same illumination conditions by Hamamatsu large area photodiode whose spectral response is given by the manufacturer. The optical properties of the materials used were measured using a UV/visible spectrometer (SAFAS 200 DES).

3. RESULTS AND DISCUSSION

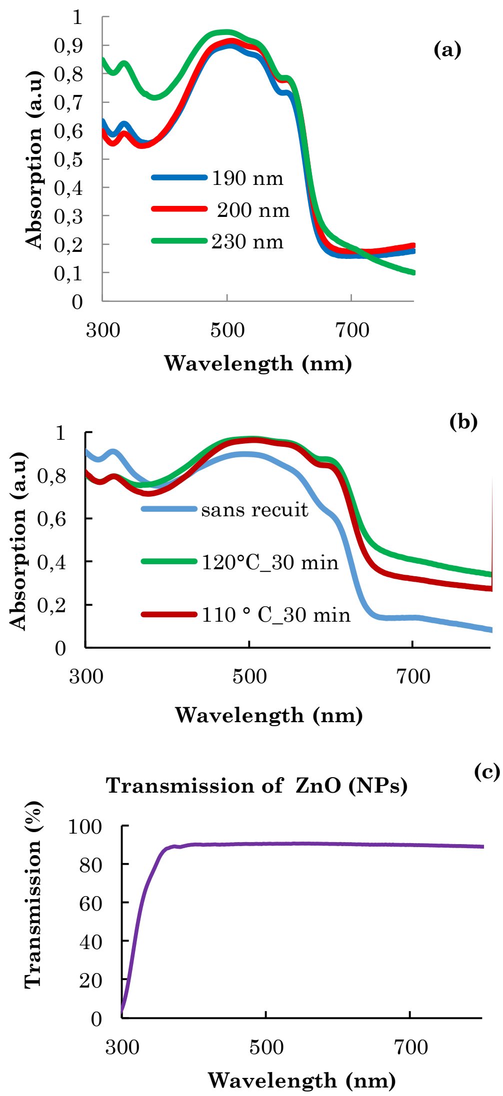
3.1 Optical Properties of the Materials Used in the Elaboration of the IOSCs

The absorption spectra of P3HT:PCBM films with varied thicknesses are presented in Fig. 1a; results show that the absorption increases as the layer thickness increases.

Annealing treatment influences the organic photovoltaic device efficiency significantly. Generally, it is believed that annealing can improve the crystallization and orientation of polymer, and it is found that the nanoscale phase separation occurs between polymer and fullerene after the thermal self-organization. Under the thermal annealing processing, the refractive index (n) and extinction coefficient (k) of the active layer varies with different temperatures. Fig. 1b shows the absorption spectra of P3HT:PCBM (1:0.8) ratio for different annealing temperatures. The results show that when the temperature increases the absorption increases.

Optical absorption of thin films of a blend of P3HT:PCBM with an approximate thickness of 190 nm is investigated. The peak absorption of P3HT increases upon thermal annealing as is evident in Fig. 1b. On the contrary to P3HT, the peak absorption intensity of PCBM reduces upon thermal annealing. This is attributed to the diffusion of PCBM molecules forming needle like crystals and subsequently at higher temperatures forming clusters [13].

The ZnO NPs were used as a buffer layer (ETL) to enhance electron transport between the cathode (ITO) and the active layer. Fig. 1c shows the transmission of ZnO layer (30 nm) used in the fabrication of the BHJ-IOSCs devices; it is about 90 % in the visible spectrum.



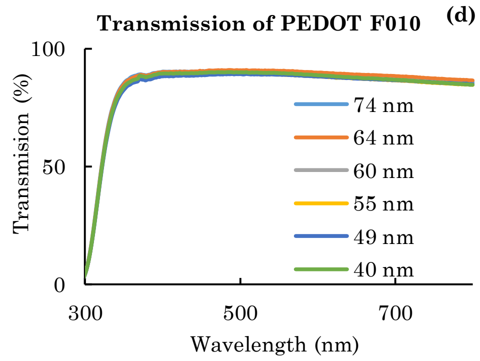


Fig. 1 – Optical properties of the materials used in the elaboration of the IOSCs: (a) Absorption of P3HT:PCBM as a function of the thickness, (b) Absorption depending on the annealing temperature over a spectrum range of 300-800 nm, (c) Transmission of ZnO NPs (30 nm) annealed at 130 °C for 15 min and (d) Transmission of PEDOT(F010) as a function of the thickness

PEDOT (F010) was used to facilitate hole transport between the active layer and the anode (Ag). Fig. 1d shows the dependence of the transmission on the PEDOT thickness. In this study, the PEDOT thickness used in the fabrication of cells is 40 nm. The transmission of PEDOT (F010) in the visible light is about 90 %.

3.2 Photovoltaic Performances

In order to evaluate the effect of the active layer thick-ness *d*, we manufactured several inverted organic solar cells. The structure of the cells is shown in Fig. 2. The cells were prepared with 190, 200 and 230 nm of P3HT:PCBM active layer, keeping the other thicknesses of buffer layers constant. The current-voltage characteristics of the cells are shown in Fig. 3. The electrical parameters of organic cells are shown in Table 1. Results show that the best conversion efficiency is obtained for a thickness of 200 nm of P3HT:PCBM layer, $\eta = 4.36$ %. This is due to the decrease of the series resistance R_s, the increase of the shunt resistance R_{sh} and the increase of the fill factor FF (FF = 66 %).

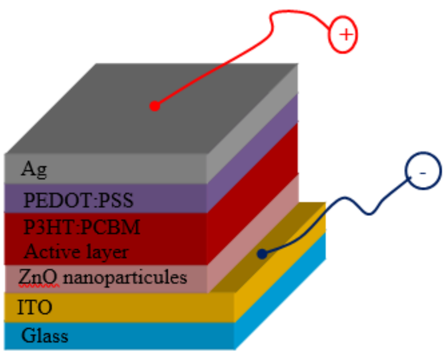


Fig. 2 – Schematic representation of the structure of the inverted organic solar cells Glass/ITO/ZnO NPs/P3HT:PCBM/PEDOT(F010)/Ag

The spectral response indicates that photons that are more strongly absorbed by P3HT are less likely to be converted into collected charge carriers, as observed in planar heterojunction devices with a layer thickness much greater than exciton diffusion length [8]. The active layer thickness has been limited largely to 100-200 nm, although a thicker active layer should absorb more incident light to be more efficient in power conversion. Thick-film polymer BHJ-OPV devices, however, have been found to be less efficient than thin-film devices because of inadequate mobility for supporting charge transport to electrodes for collection [9, 10].

External quantum efficiency (EQE) represents the ration between the measured photocurrent and the intensity of the incoming monochromatic light. Fig. 4 shows the EQE of the IOSCs as a function of the active layer thickness.

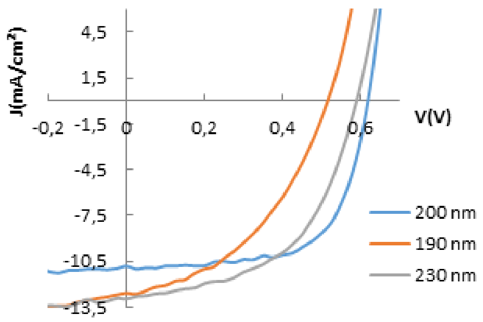


Fig. 3 – $J(V)$ characteristics of the cell as a function of the active layer thickness

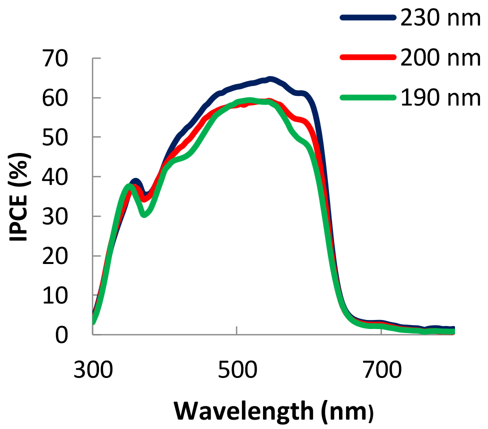


Fig. 4 – IPCE of the inverted OSCs as a function of the P3HT:PCBM active layer thickness

4. INFLUENCE OF LIGHT INTENSITY

The thicknesses of glass/ITO/ZnO (np)/P3HT:PCBM/PEDOT (F010) inverted cell layers are as follows: ZnO (30 nm), P3HT:PCBM (200 nm) and PEDOT (40 nm). The cells were annealed in the glove box at 110 °C for 30 min. The voltage current density characteristics in Fig. 5 were measured for different luminous intensity values in the glove box, 1 sun corresponds to AM 1.5 (100 mW/cm^2).

Table 2 shows the electrical parameters of the glass/ITO/ZnO (np)/P3HT:PCBM/PEDOT:PSS/Ag as a function of the light intensity. The cells produced have an area of 0.18 cm^2, the characterizations are carried out under different light intensities with 1 sun =100 mW·cm^{-2}. Results show that the efficiency of an organic photovoltaic cell increases as the light intensity increases. The voltage

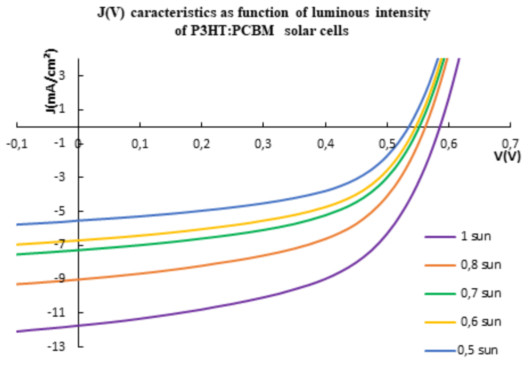


Fig. 5 – Dependence J(V) of a P3HT:PCBM organic cell as a function of luminous intensity

V_{OC} varies linearly with the light intensity. Our results show that: $V_{OC} = 0.1 * light\ intensity + 0.47$. The short circuit current density J_{sc} and the efficiency η increase with increasing light intensity, J_{sc} = 5.52 mA/cm², V_{oc} = 0.52 V and η = 1.52 % under light intensity of 0.5 sun, J_{sc} = 11.73 mA/cm², V_{oc} = 0.57 V and η = 3.65 % under light intensity of 1 sun.

5. EFFECT OF THE ANNEALING TEMPERATURE

Thermal annealing improves the conversion efficiency of organic cells. When the temperature increases, the yield increases as shown in Fig. 6. The efficiency of the devices without annealing is 2.26 %. After annealing the efficiency η increases: at 100 °C, η = 2.26 %; at 125 °C, η = 3 %; at T = 150 °C, η = 3.84 %. Above 150 °C, the yield decreases: at 175 °C, η = 3.56 %. Under annealing treatments the polymer is seen to be ordered and devices with ordered structures show higher efficiency [16]. An efficiency η = 0.52 % is obtained for cells without annealing and after annealing of the device η = 0.78 % [17]. In this work, for temperatures above 150 °C, the yield decreases as shown in Fig. 6, this is due to the degradation of organic materials under high temperature. Under higher temperatures, there is a threshold temperature T_{max}, at which organic solar cells become thermally unstable [18].

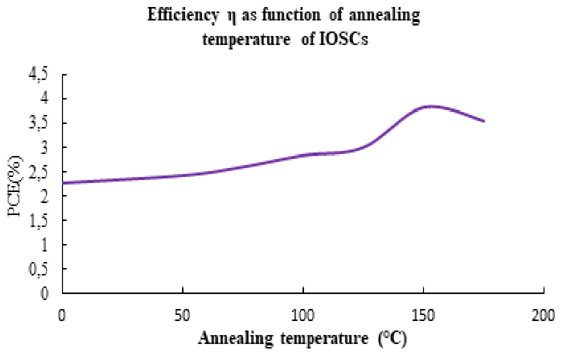


Fig. 6 – Effect of the thermal annealing temperature on the performances of IOSCs

Table 1 – Electrical parameters of BHJ-IOSCs devices measured under AM 1.5 illumination with varying active layer thickness

Active layer thickness (nm)	V_{OC} (V)	J_{SC} (mA/cm²)	FF (%)	PCE (%)	Rs (Ω)	Rsh (Ω)
190	0.58	12.85	53	3.98	48.52	1656.27
200	0.60	10.95	66	4.36	31.32	4981.73
230	0.60	11.76	59	4.21	34.88	1990.85

Table 2 – Electrical parameters of the organic solar cells under different light intensities

Light intensity	1 Sun	0.8 sun	0.7 sun	0.6 sun	0.5 sun
V_{oc} (V)	0.57	0.55	0.54	0.53	0.52
J_{sc} (mA.cm^{-2})	11.73	8.97	7.25	6.70	5.52
FF	0.54	0.54	0.54	0.53	0.53
ɂ (%)	3.65	2.65	2.09	1.90	1.52
P_{max} (mW·cm^{-2})	3.65	2.65	2.09	1.90	1.52
R series (Ω)	52.75	64.19	67.55	66.28	93.19
R shunt (Ω)	1421.99	1723.80	1978.41	2052.58	2246.44

6. CONCLUSIONS

The performances of IOSCs depend on the active layer thickness and annealing temperature. We established that the optical absorption intensity for P3HT:PCBM layers enhances with thickness enhancement. The optical absorption intensity for PCBM parts of the spectrum reduces with annealing temperatures. In this work, we get a maximum efficiency of 4.36 % with a 200 nm of P3HT:PCBM layer. From this work, we can say that optimization of the layer thickness is a promising factor for high efficiency of IOSCs.

The conversion efficiency of organic solar cells increases as the light intensity increases. This is due to the increase of the short-circuit Jsc and the increase of the open circuit voltage Vco. The series resistance and the shunt resistance decrease when the light intensity increases. However, the higher the luminosity, the more a cell converts solar energy into electricity.

Thermal annealing improves the efficiency of the cells this is due to the improvement of the crystallinity and the morphology of the organic layers and to the good adhesion of the organic layers with the electrodes. If the annealing temperature exceeds 150 °C, the efficiency decreases because of the degradation of the plastic materials under high temperatures.

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Залежність продуктивності органічних сонячних елементів як функції товщини активного шару, світлової інтенсивності та температури теплового відпалу

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Фотоелектричні елементи це оптико-електронні пристрої для перетворення світлової енергії в електричну. У роботі досліджено вплив товщини активного шару на продуктивність інвертованих сонячних елементів. Структурами виготовлених пристроїв є Glass/ITO/ZnO NPS/P3HT:PCBM/PEDOT (FO10)/Ag. Активний шар на основі суміші полі (3-гексилтіофен) і фулерену із співвідношенням (1:0.8) є спін-покриттям з різною швидкістю отримання різних товщин. ZnO NPS (30 нм) використовували для поліпшення транспорту електронів між ITO і активним шаром. PEDOT (F010) (40 нм) використовували як дірковий транспортний шар. Комерційний ITO, використаний у цьому дослідженні, має опір листа 7 Ω/□ при товщині 110 нм. У роботі описано виготовлення та характеристики комірок, а також оптичні властивості використаних матеріалів та фотоелектричні параметри сонячних елементів. Обґрунтовано прямий зв'язок між товщиною активного шару та ефективністю інвертованих сонячних елементів. Для 200 нм P3HT:PCBM ефективність складає 4.36 %. Обговорено вплив інтенсивності світла та термічного відпалу на продуктивність фотоелектричних елементів. Характеристики клітин показують, що інтенсивність світла впливає на електричні та оптичні параметри органічних клітин.

Ключові слова: Перевернуті органічні клітини, Поглинання, IPCE, Товщина, Інтенсивність світла, Термічний відпал.