

Double ETL in ITO-free poly-3-hexylthiophene-based organic solar cells

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Abstract— Energy level and power conversion efficiency were investigated in the inverted organic solar cells with configuration FTO/TiO₂/P3HT:PC₆₁BM/MoO₃/Ag and FTO/PFN/P3HT:PC₆₁BM/MoO₃/Ag structures in a double electron transport layer (ETL) organic solar cell with configuration FTO/TiO₂/PFN/P3HT:PC₆₁BM/MoO₃/Ag. The influence of TiO₂ and PFN as ETL was evaluated individually and together, in solar cells and an UV-Vis experiment, showing that the use of a double ETL can increase the efficiency up to 2.5 times compared to cells where only a single layer was used. The best cell built with double ETL showed a maximum efficiency of 4.29%.

Index Terms— organic solar cell; transport layer; double ETL.

I. INTRODUCTION

Although organic solar cells (OPV) have been studied since the 1970s (with efficiencies on the order of ~10⁻⁴% [1]) in recent years, they have been of great interest to scientists as they are considered relatively inexpensive to manufacture, friendly to the environment as it lacks inorganic components that are harmful to the planet, due to its excellent electrical, optical, thermal and mechanical properties, for presenting substantial increases in efficiency in recent years and for being, apparently so far, the only type of technology capable of addressing large-scale manufacturing through roll-to-roll processes. [2]–[4]. However, OPVs still have low efficiencies compared to inorganic cells, so research and development of this type of technology should be further encouraged.

Among the existing OPVs, in the active layer, the use of P3HT polymers, as donor material, and PC₆₁BM, as acceptor material, has been extended, due to its excellent optical and electrical properties, as well as for presenting an excellent stability and lifetime [5], [6]. In addition, transport layers, a hole transport layer (HTL) and an electron transport layer (ETL), are typically incorporated between the active layer and the electrodes to increase power conversion efficiency (PCE) [7], [8]. These transport layers have the function of allowing the passage of sunlight, extracting and collecting photo-generated charge carriers, reducing their recombination, reducing surface imperfections and improving stability in air and life time [7]–[13].

In this research, the use of molybdenum trioxide (MoO₃) as a hole-transporting layer was proposed, while titanium dioxide (TiO₂) and the Poly [(9,9-bis(3'-(N,N-

dimethylamino)propyl)-2,7-fluorene)-alt-2,7-(9,9-dioctylfluorene)] (PFN) function as an electron-transporting layer and a buffer layer, respectively, although formally the latter is also considered an ETL. Both MoO₃ and TiO₂ were chosen for being considered promising, in addition to presenting excellent mobility, stability, low cost and high transparency [14]–[16]. Meanwhile, PFN is speculated that it improves the interfacial contact between PC₆₁BM and TiO₂, facilitating the efficient transport and collection of electrons [17]. Fluorine-doped tin oxide (FTO) was used as a transparent conductive oxide due to its relative low cost, compared to indium tin oxide (ITO), its excellent electrical properties, high mobility and mechanical stability [18]–[20]. In addition, it shows great thermal stability over 350 °C, unlike ITO, which allows the TiO₂ to be deposited and thermally treated to reach the Anatase phase [19].

Here, the details of the experiment carried out are shown, the UV-Vis spectra of TiO₂, PFN and TiO₂/PFN layers are shown and subsequently the devices with configuration FTO/TiO₂/P3HT:PC₆₁BM/MoO₃/Ag, FTO/PFN/P3HT:PC₆₁BM/MoO₃/Ag and FTO/TiO₂/PFN/P3HT:PC₆₁BM/MoO₃/Ag that were manufactured and compared to determine the efficiency of using the electron-transporting layers, TiO₂ and PFN, individually and together.

II. EXPERIMENTAL DETAILS

FTO substrates (Pilkington Tec15) were consecutively cleaned in an ultrasonic bath using soap, acetone, and ethanol for 15 minutes each one. TiO₂ sol-gel solution was prepared under nitrogen atmosphere using titanium (IV) isopropoxide (Sigma-Aldrich, 97%), ethanol (J.T. Baker, 99.8%) and hydrofluoric acid (J.T. Baker, 98.51%). Firstly, 0.117 mL of hydrofluoric acid was added into 14.2 mL of ethanol and then 4 mL of titanium (IV) isopropoxide was added into 14.2 mL of ethanol in a different vial. Finally, both solutions were mixed and stirred for 5 minutes. After that, the solution was spin-coated onto the glass and FTO substrates (for UV-Vis and cells experiments, respectively) and annealed at 550 °C for 1 hour to obtain a thickness of ~70 nm. PFN's solution was made using 1 mL of methanol with the addition of 2% by volume of acetic acid to make a solution of 2 mg/mL of PFN and it was deposited using spin-coating (onto the glass and FTO substrates) and annealed at 100 °C for 8 min to obtain a thickness of ~10 nm. Besides, active layer blend was made by

dissolving P3HT:PC₆₁BM (SOL4106 and SOL5061A, respectively, Solaris Chem, 99.5%) at 1:1 w/w, 25 mg/mL in o-dichlorobenzene (J. T. Baker, 98.5%) under the nitrogen atmosphere, and the solution was stirred at 60°C for 30 minutes. Then, the blend was deposited by spin coating and annealed at 100 °C for 8 min to obtain a thickness of ~250 nm. After that, the MoO₃ layer (~10nm) was deposited by physical vapor deposition (PVD) using MoO₃ powder (Sigma-Aldrich 99.5%). Finally, silver (Ag) metallic contacts with a thickness of ~100 nm were deposited by PVD using a mask with a nominal diameter of 1mm. Finally, Current density-voltage (J-V) curves were obtained from the solar simulator model LCS-100 calibrated with AM 1.5.

III. RESULTS AND DISCUSSIONS

Fig. 1 shows the energy level diagram of the organic solar cells constructed with configuration a) FTO/TiO₂/P3HT:PC₆₁BM/MoO₃/Ag, b) FTO/PFN/P3HT:PC₆₁BM/MoO₃/Ag and c) FTO/TiO₂/PFN/P3HT:PC₆₁BM/MoO₃/Ag. It is observed that both TiO₂ and PFN present ideal characteristics to be used in this structure, since titanium dioxide has a conduction band of -4.2 eV [16], [21], while PFN presents a work function of -4.2 eV [22], [23], as well. This allows proper transport of the negative charge carriers from the lowest unoccupied molecular orbital (LUMO) of the PC₆₁BM (-3.9 eV) [6], [24], [25], to the transparent conductive oxide (-4.4 eV) [26]. The energy difference at the interface (ΔE_i) PC₆₁BM/PFN, for the structure of Fig. 1 - b) and c), and PC₆₁BM/TiO₂, in the case of structure a), is equal to -0.3 eV. While the energetic difference between PFN/FTO, of structure b), and TiO₂/FTO, of structure a) and c), is -0.2 eV. Finally, there is no energetic difference in the PFN/TiO₂ interface, in structure c), therefore $\Delta E_i = 0$ eV. Continuing with the analysis of Fig. 1, it is observed that the active layer, composed of the polymers P3HT and PC₆₁BM, show an energy difference of the lowest unoccupied molecular orbital ($\Delta LUMO$) of -0.6 eV [6], [24], [25], this being enough energy to dissociate a exciton in an electron-hole pair whose charges will be transported towards the anode and cathode and with the help of the transport layers. Furthermore, the difference between the highest occupied molecular orbital of the donor polymer (HOMO) and the LUMO of the acceptor polymer is equal to 1.3 eV whose value is considered to represent the maximum theoretical open circuit voltage (V_{oc}) that a solar cell can reach with this active layer, however, it can be verified that the maximum V_{oc} in practice is close to 0.6 V [27]. Finally, the work function of MoO₃ (-5.3 eV) has a lower energy than the HOMO of P3HT (-5.2 eV), in addition to there is a wide gap between this and the work function of Ag (-4.4 eV) [28] and therefore there is a probability of recombination in these interfaces, however, the transport mechanism of the charge carriers of this HTL is by tunneling thanks to its low thickness (~10 nm), in addition to the excellent extraction and transport of negative charge carriers due to ETL, greatly reducing the probability of recombination [29].

Fig. 2 shows the transmission spectra of TiO₂ and PFN thin films and the combination of both layers (TiO₂/PFN). TiO₂ transmission starts around 300 nm and increase from 58% to over 90% after 400 nm and transmission and PFN transmission goes from 60%, at 400 nm, to over 97% at 450 nm. Also, PFN has an absorption range of 300 nm to 450 nm. Meanwhile, TiO₂/PFN transmission was measured to know the influence of the PFN layer on the TiO₂ layer and it shows a change in the normal TiO₂ transmission spectra due to the

strong influence of the PFN absorption in the range of 300 nm to 450 nm. In fact, the light transmitted by the TiO₂ layer is absorbed by the PFN layer, so the transmission in general decreases in contrast to individually deposited layers.

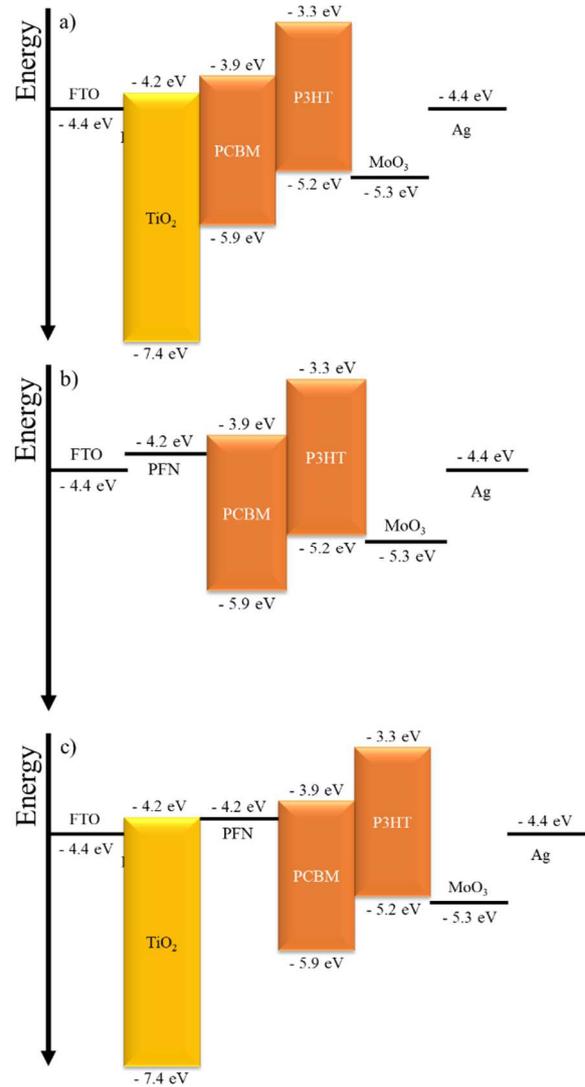


Fig. 1. Energy level diagram of structures a) FTO/TiO₂/P3HT:PC₆₁BM/MoO₃/Ag, b) FTO/PFN/P3HT:PC₆₁BM/MoO₃/Ag and c) FTO/TiO₂/PFN/P3HT:PC₆₁BM/MoO₃/Ag.

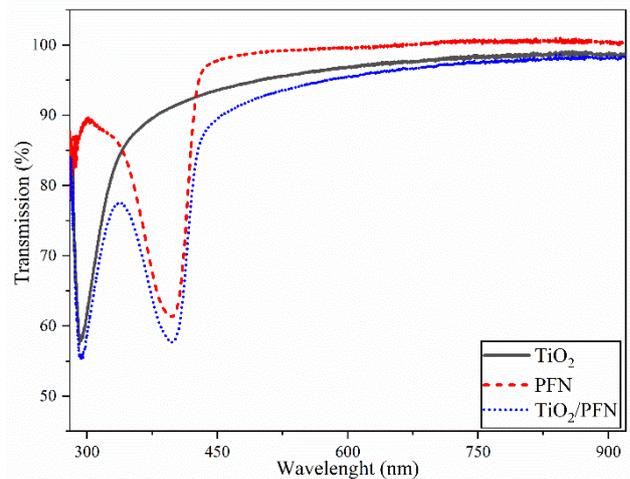


Fig. 2. Transmission spectra of TiO₂, PFN, and TiO₂/PFN layers.

In Fig. 3 we can see Tauc's plots of TiO₂ and TiO₂/PFN layers. In part *a*) TiO₂ shows a band gap (E_g) of 3.23 eV, which is approximately the normal indirect band gap found in literature for this material [21]. Fig. 3 *b*) shows the TiO₂/PFN Tauc plot, where we can see a fold corresponding to the absorption range of PFN and after that the slope corresponding to TiO₂ with which an indirect bandwidth of 3.12 was calculated. Considering that E_g is the minimum energy necessary to excite an electron from its bound state to a free state, using double ETL could reduce the energy required to transport the electrons. It's important to mention that E_g 's PFN cannot be calculated accurately with this method and therefore the corresponding plot is omitted.

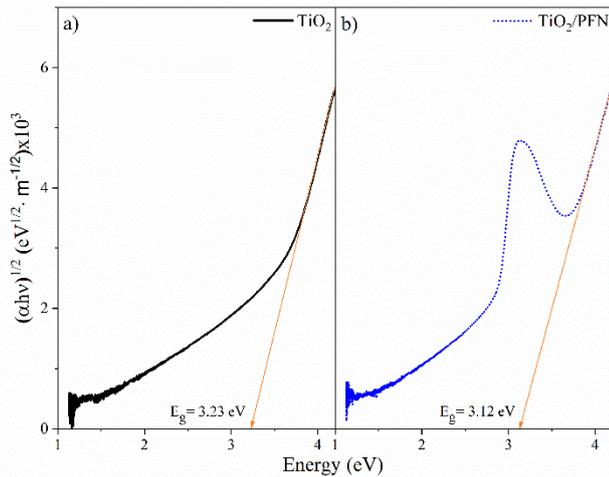


Fig. 3. Tauc's plot of a) TiO₂ and b) TiO₂/PFN layers.

Table I and Fig. 4 show the results of the devices built with configuration FTO/TiO₂/P3HT:C₆₁BM/MoO₃/Ag, FTO/PFN/P3HT:PC₆₁BM/MoO₃/Ag and FTO/TiO₂/PFN/P3HT:PC₆₁BM/MoO₃/Ag. It is observed that

cells built with only the ETL of TiO₂ have a short circuit current density (J_{sc}) between 11.81 and 12.39 mA/cm², an open circuit voltage (V_{oc}) in the range of 0.28 to 0.40 Volts and reaching filling factors (FF) between 0.41 and 0.43, with which an energy conversion efficiency (PCE) was obtained between 1.35% and 1.97% and with an average efficiency (Av. PCE) of 1.59%. Meanwhile, when the PFN is used as ETL, a J_{sc} oscillating between 11.15 and 11.44 mA/cm² was observed, a V_{oc} between 0.38 and 0.42 Volts and a FF in the interval of 0.32 to 0.35, with which a PCE that goes from 1.46% to 1.54% and with an Av. PCE of 1.52%. Finally, the device manufactured with double ETL (FTO/TiO₂/PFN/P3HT:PC₆₁BM/MoO₃/Ag) presented a minimum J_{sc} of 11.99 mA/cm² and a maximum of 13.01 mA/cm², reaching a V_{oc} of 0.60 Volts and an FF oscillating between 0.43 and 0.55, thus obtaining a minimum solar efficiency of 3.14% and a maximum PCE of 4.29%, with an average of 3.90%.

TABLE I. ELECTRICAL PROPERTIES OF THE CONSTRUCTED CELLS USING TiO₂, PFN AND TiO₂/PFN AS TRANSPORT LAYERS.

ETL	Sample	J_{sc} (mA/cm ²)	V_{oc} (Volts)	FF	PCE (%)	Av. PCE (%)
TiO ₂	1	11.97	0.28	0.41	1.35	1.59
	2	11.92	0.40	0.42	1.97	
	3	11.81	0.28	0.42	1.37	
	4	12.39	0.33	0.43	1.77	
	5	12.04	0.30	0.41	1.49	
TiO ₂ /PFN	1	13.01	0.60	0.55	4.29	3.90
	2	12.19	0.60	0.43	3.14	
	3	12.47	0.60	0.54	4.06	
	4	12.51	0.60	0.54	4.10	
	5	11.99	0.60	0.55	3.93	
PFN	1	11.35	0.42	0.32	1.54	1.52
	2	11.15	0.40	0.33	1.46	
	3	11.44	0.39	0.34	1.55	
	4	11.30	0.40	0.33	1.52	
	5	11.29	0.38	0.35	1.53	

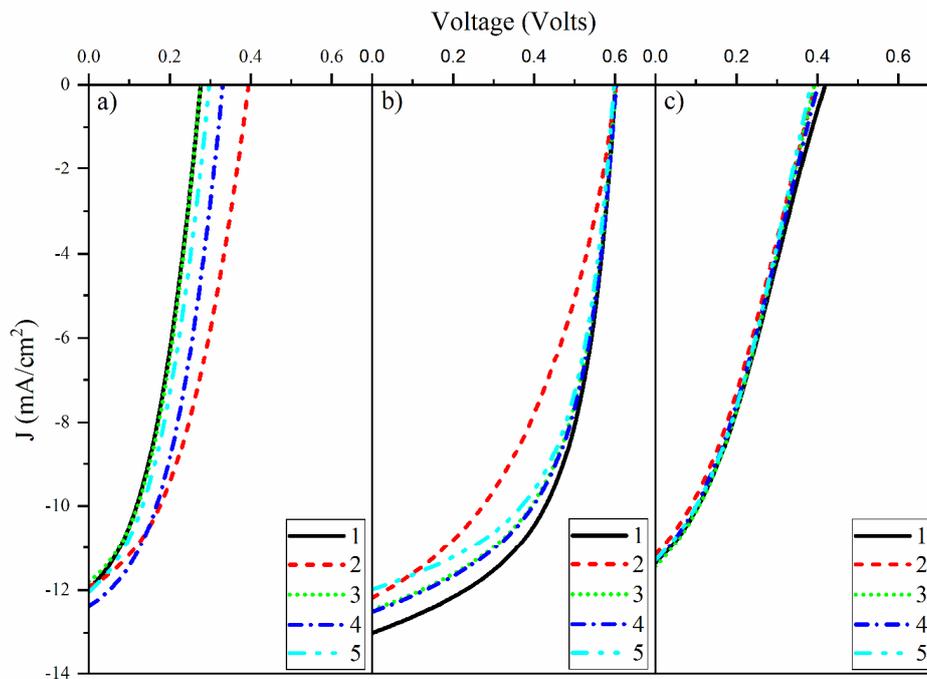


Fig. 4. J-V curves of organic solar cells with structure configuration a) FTO/TiO₂/P3HT:C₆₁BM/MoO₃/Ag, b) FTO/TiO₂/PFN/P3HT:C₆₁BM/MoO₃/Ag and c) FTO/PFN/P3HT:C₆₁BM/MoO₃/Ag.

PFN and TiO₂, in this experiment, showed very similar energy conversion efficiency, which is far exceeded when both layers are combined in the same device. In fact, this average is equivalent to 245% of the efficiency obtained when only TiO₂ is used as ETL and equivalent to 256% with respect to the efficiency calculated when only PFN is used as ETL, that is, in both cases, practically 2.5 times more efficient use of double ETL. Due to its polymeric properties, the PFN is in tune with the PC₆₁BM, greatly increasing the interfacial contact surface and having a very low energy difference (-0.3 eV), efficiently extracting the negative charge carriers, which is observed in the high open circuit voltages, relative to voltages with TiO₂ as ETL. On the other hand, the TiO₂ generates an ohmic contact with the FTO when merging and reaching the Anatase phase at 550 °C, increasing the Jsc and the FF, with respect to the PFN. By using both ETLs, the benefits found in these are added and, since the TiO₂/PFN interface does not generate an energy difference, electrons can flow more efficiently from the acceptor polymer to the transparent conductive oxide (FTO).

Today, as far as is known, there is no cell with a double carrier layer (TiO₂/PFN) in this type of structure (FTO/TiO₂/PFN/P3HT:PC₆₁BM/MoO₃/Ag), so it could not be compared so easily, however, it can be compared with a P3HT:PC₆₁BM base cell whose PCE obtained in the laboratory is usually around 5%, so Table II shows a comparison of the best cells built against a cell base with configuration ITO/PEDOT:PSS/P3HT:PC₆₁BM/Al, whose Jsc is 12.80 mA/cm², a Voc of 0.64 Volts, a FF of 0.63 and efficiency of 5.15% [30]. Meanwhile, the best cell built in this work was with the configuration FTO/TiO₂/PFN/P3HT:PC₆₁BM/MoO₃/Ag, whose Jsc is 13.01 mA/cm², with a Voc of 0.60 Volts, FF of 0.55 and an efficiency of 4.29%. Comparing both efficiencies it could be said that a determining factor is the fill factor, indicative that there are series and parallel resistances in the constructed cell, possibly due to the existing recombination probability in the HTL.

TABLE II. ELECTRICAL PROPERTIES OF THE BEST CONSTRUCTED CELLS USING TiO₂, PFN AND TiO₂/PFN AS TRANSPORT LAYERS AGAINST A TYPICAL ORGANIC SOLAR CELL.

Structure configuration	Jsc (mA/cm ²)	Voc (Volts)	FF	PCE (%)
FTO/TiO ₂ /AL/MoO ₃ /Ag	11.92	0.40	0.42	1.97
FTO/PFN/AL/MoO ₃ /Ag	11.44	0.39	0.34	1.55
FTO/TiO ₂ /PFN/AL/MoO ₃ /Ag	13.01	0.60	0.55	4.29
ITO/PEDOT:PSS/AL/Al*	12.80	0.64	0.63	5.15

AL: Active layer (P3HT:PC₆₁BM).

* Reference [30]

IV. CONCLUSIONS

UV-Vis spectra shows that TiO₂ layer decrease its Eg, by using a PFN layer, from 3.23 eV to 3.12 eV reducing the energy required to transport the electrons. Cells built with FTO/TiO₂/P3HT:PC₆₁BM/MoO₃/Ag, FTO/PFN/P3HT:PC₆₁BM/MoO₃/Ag and FTO/TiO₂/PFN/P3HT:PC₆₁BM/MoO₃/Ag configuration showed good electrical properties. While PFN like ETL manages to increase open circuit voltage, TiO₂ has a considerable impact on short circuit current density and filling factor. In addition, when using a double ETL composed of TiO₂/PFN, the benefits of both layers are complemented by increasing energy conversion efficiency almost 2.5 times, compared to using only one of them. The best cell built has a configuration FTO/TiO₂/PFN/P3HT:PC₆₁BM/MoO₃/Ag with

a Jsc of 13.01 mA/cm², a Voc of 0.60 Volts, an FF of 0.55 and an power conversion efficiency of 4.29%. The possibility exists of improving this efficiency by decreasing the probability of recombination of the charge carriers by choosing a different HTL or metallic contact.

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