Effect of Hole Transport Layers on the Performance of Organic Optoelectronic Devices based on PBDB-T:ITIC Bulk Heterojunction

Shan-Shan ZHANG, Xiao-Hua ZHANG and Jiang HUANG*

State Key Laboratory of Electronic Thin Films and Integrated Devices, School of Optoelectronic Information, University of Electronic Science and Technology of China (UESTC), Chengdu 610054, P.R. China

*huangjiang@uestc.edu.cn

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Abstract. In this work, the organic optoelectronic devices with both photovoltaic and detection performance were fabricated based on a blend of the polymer poly[(2,6-(4,8-bis(5-(2-ethylhexyl)thiophen-2-yl)-benzo[1,2-b:4,5-b']dithiophene))-alt-(5,5-(1',3'-di-2-thienyl-5',7'-bis(2-ethylhexyl)benzo [1',2'-c:4',5'-c']dithiophene-4,8-dione))] (PBDB-T) with the non-fullerene acceptor of 3,9-bis(2-methylene-(3-(1,1-dicyanomethylene)-indanone)-5,5,11,11-tetrakis(4-hexylphenyl)-dithieno [2,3-d:2',3'-d']-s-indaceno[1,2-b:5,6-b'] dithio-phenylene) (ITIC). Meanwhile, the different hole transport layers (HTL) materials of N,N'-bis-(1-naphthyl)-N,N'-diphenyl-1,1'-biphenyl-4,4'-diamine (NPB) and N,N'-diphenyl-N,N'-bis(3-methylphenyl)-(1,1'-biphen-yl)-4,4'-diamine (TPD) were adopted to modify the interface of organic active layer and Ag/MoO₃ electrode. The result showed that without thermal annealing of PBDB-T:ITIC, the photovoltaic device with TPD HTL exhibited an improvement in power conversion efficiency (PCE) from 4.45% to 5.26% compared with the control device without HTL. Moreover, by analyzing the dark current behavior after thermal annealing, it was found that the TPD HTL could effectively suppress the leakage current from Ag/MoO₃ electrode to the active layer. As a result, an efficient organic photo detector with a detectivity of 3.87×10^{10} Jones was achieved.

Introduction

Over the past decades, most bulk-heterojunction (BHJ) organic optoelectronic devices use fullerene derivative materials as the electron acceptors and yield high photovoltaic and detective performance with advantages of light weight, flexibility and low-cost [1-3]. However, fullerene materials have disadvantages such as weak absorption of sunlight and low tunability of electronic energy levels. Those limitations offer a rapid developmental opportunity of non-fullerene acceptor materials in organic optoelectronic device [4-6]. Based on the non-fullerene material ITIC, an outstanding power conversion efficiencies (PCEs) higher than 11% was achieved by S. Li in the organic photovoltaic (OPV) [7]. Zhan and co-workers had reported organic photo detector (OPD) with a high detectivity (D^*) of 10^{12} Jones at ± 15 V, because the band bending decreases the tunneling-injection barriers of the oppositely charged carriers under light illumination [8]. However, the OPD based on photovoltaic effect using the PBDB-T : ITIC active layer has not been reported. In this work, we fabricated organic optoelectronic devices with photovoltaic and detective performance based on the active layer of PBDB-T:ITIC. Also, the NPB and TPD were employed as hole transport layers (HTL) to improve the PCE of OPV and reduce the dark current of OPD.

Experimental

The molecular structures of ITIC, PBDB-T, TPD and NPB are shown in Fig. 1(a), and the schematic device structure is indium tin oxide (ITO)/ZnO (40 nm)/PBDB-T:ITIC (100 nm)/HTL (10 nm)/ MoO₃ (15 nm) /Ag (100 nm) as depicted in Fig. 1(b). The ITO-coated glass substrates with a 10 Ω /sq sheet resistance were cleaned successively in an ultrasonic bath containing detergent, acetone, deionized water, and isopropyl alcohol each step for 15 min, and dried in an oven for 2h at

90°C [9]. Prior to spin-coating, the substrate was treated with UV light for 10 min. ZnO nanoparticles was spin-coated at 5000 r.p.m. for 40 s on the ITO-glass substrates. The PBDB-T was blended with ITIC (ratio = 1:1 by weight) and a concentration of 20mg/ml was dissolved in chlorobenzene (CB) with the addition of 0.5% 1,8-diiodooctane (DIO). Then the solution was spin-coated onto the ZnO at 2500 r.p.m. for 60 s and the thickness was about 100 nm. Then NPB and TPD were deposited as HTLs at a rate of 1 to 2 Å/s at a pressure of 3.0×10^{-4} Pa, followed by the deposition of MoO₃ (15 nm) at a rate of 0.1 Å/s. 100 nm thick Ag electrode was thermally vacuum deposited (1Å/s, 2×10^{-3} Pa).

Fig. 1(a) Chemical structures of ITIC, PBDB-T, NPB and TPD. (b) Device structure in this work. (c)Energy band diagram of materials used OPD.

Result and Discussion

Performance of OPV with Different HTLs

Fig. 2. J–V characteristics of OPV with and w/o HTLs under dark and 100 mW/cm² irradiation.

Fig. 2 illustrates the effect of different HTLs on J-V characteristics of OPV. The detailed parameters including open circuit voltage (V_{oc}), Short circuit current (J_{sc}), fill factor (FF), and power conversion efficiency (PCE) are listed in Table 1. Without thermal annealing of PBDB-T : ITIC, the device without a HTL showed a V_{oc} of 0.85 V, J_{sc} of 11.08 mA/cm², FF of 47.45% and PCE of 4.45%. By utilizing TPD as a HTL, an improvement on the V_{oc} of 0.87V, J_{sc} of 12.02 mA/cm² and FF of 50.52% were obtained, which leads to the enhancement of PCE to 5.26%. However, the NPB doesn't have an enhancement on the device performance. This remarkable improvement results from that TPD has compensatory absorption from 400 to 550 nm, while the absorption spectrum of NPB mainly distributed in the UV region (see Fig. 3.) and the hole mobility $\sim 1.0 \times 10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ of TPD is much higher than that $\sim 9.0 \times 10^{-4} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ of NPB [10-12].

Fig. 3. UV-vis absorption spectra of pristine films of ITIC, PBDB-T, NPB, and TPD.

Performance of OPDs with Different HTLs

Fig. 4 displays the characteristics of current density versus the voltage bias of OPDs with different HTLs in log-scale, and the photo detective performances were summarized in Table 1. It can be seen that the dark current density (J_{dark}) of devices with TPD HTL has been decreased by one order of magnitude than the control device without TPD HTL after thermal annealing (TA) at 160°C , indicating that the inserted HTL between the active layer and MoO_3/Ag anode is beneficial to reduce the reversed dark current. At the bias of -0.5V , the $I_{\text{on}}/I_{\text{off}}$ of the TA treated OPD w/o a HTL is 176.33, while the untreated control device has a better $I_{\text{on}}/I_{\text{off}}$ of 354.84. It is worth noting that the highest performance improvement with $I_{\text{on}}/I_{\text{off}}$ of 387.12 and detectivity D^* of 3.87×10^{10} Jones using 5 nm TPD as a HTL can be obtained. While without thermal annealing of PBDB-T:ITIC, the NPB and TPD HTLs have no effect on the overall performance of the OPDs. This performance improvement of OPDs originates from two important aspects. One is that the TPDHTL can effectively block electron injection from the MoO_3/Ag anode because of the high electron injection barrier of 3.4eV (see Fig 1.1(c)), and the other is the lower electron mobility of TPD than NPB[13-15].

Fig.4. J-V characteristics of OPDs with and w/o HTLs under dark and $100\text{ mW}/\text{cm}^2$ irradiation.

Table 1. The device characteristics of organic optoelectronic devices based on PBDB-T:ITIC active layer with and w/o different HTLs.

Device	V_{oc} (V)	J_{sc} (mA/cm^2)	FF(%)	PCE (%)	J_{dark} (A/cm^2)	$I_{\text{on}}/I_{\text{off}}$	D^* (Jones)
w/o+TA	0.83	11.34	51.00	4.79	1.00×10^{-4}	176.33	3.10×10^{10}
w/o	0.85	11.08	47.45	4.45	3.50×10^{-5}	354.84	3.70×10^{10}
NPB	0.79	9.15	43.38	3.13	8.25×10^{-5}	116.22	1.85×10^{10}
TPD	0.87	12.02	50.52	5.26	9.44×10^{-5}	137.93	2.35×10^{10}
TPD+TA	0.75	10.92	43.85	3.61	3.21×10^{-5}	387.12	3.87×10^{10}

TA: thermal annealing.

Conclusions

In summary, the effect of NPB and TPD as the HTLs on the performance of organic optoelectronic device was investigated and compared. A maximum PCE up to 5.26% was obtained without thermal annealing of active layer and by incorporating thermal annealing at 160°C, leading to a high D^* of 3.87×10^{10} Jones compared to that of the control device w/o HTLs. All these findings revealed that using TPD HTL is an effective way to manufacture highly efficient organic optoelectronic device with photovoltaic and detection performance.

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