



Improving spray coated organic photodetectors performance by using 1,8-diiodooctane as processing additive

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ABSTRACT

In this work, we present and support the positive influence of 1,8-diiodooctane (DIO) as processing additive for the fabrication of solution processed near infrared (NIR) organic photodiodes (OPDs) based on the spray-coated blend of [6,6]phenyl-C70-butyric acid methyl ester (PC₇₀BM) and poly[(2,5-bis(2-hexyldecyl)-2,3,5,6-tetrahydro-3,6-dioxopyrrolo[3,4-c]pyrrole-1,4-diyl)-*alt*-([2,2':5',2''-terthioph-ene]-5,5''-diyl)] (PDPP3T). Transmission electron microscopy (TEM) analysis show that the use of DIO improves donor–acceptor interfaces in the bulk heterojunction (BHJ) layer. Besides the enhancement of electron/hole transport pathways that leads to higher efficiencies, it reduces significantly the roughness of spray coated layers. The External Quantum Efficiency (EQE) is increased up to 35% at 900 nm and the dark current is decreased one order of magnitude in comparison to the reference down to 20 nA/cm² at -5 V. The OPD's detectivity is increased from 1.66 × 10¹¹ Jones to 3.34 × 10¹² Jones and the linearity response is improved. A comprehensive morphological and electro-optical characterization of the generated layers and devices is the main focus of this work.

1. Introduction

Organic photodetectors (OPDs) offer clear advantages compared to inorganic photodetectors related to their low processing cost, the feasibility of being lightweight/flexible and the production of large active areas. Additionally, they offer the possibility to address a wide range of optical applications by tailoring the absorption spectra from the Ultraviolet (UV) – visible to the near infrared (NIR). These applications include image sensing, communications, environmental monitoring, remote control, day- and night-time surveillance, and chemical/biological sensing [1].

The big challenge for the entire organic opto-electronic community is the production of NIR sensors with a solution processed organic semiconductor that presents similar performance to silicon based pho-

todiodes. Important figures of merit include the reduction of dark current under reverse bias as well as high external quantum efficiency (EQE).

Solution processed organic electronics devices require thin films with thicknesses ranging from few tens of nm to few hundreds of nm. Spin-coating [2,3] slot-die coating and screen-printing [4] are typical deposition techniques that allow the production of thin film layers with surface roughness of few nm on large areas. However, all these techniques require flat surface substrates and the layer thickness is limited by the formulation of the solution. Thick photoactive layer thicknesses (up to 1 μm) are beneficial for the reduction of leakage currents in OPDs [5] and the techniques mentioned above can hardly achieve these thicknesses. On the other hand, spray coating technique offers the possibility to deposit any desirable thickness by multiple spray coating

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steps. Also, it is substrate and solvent independent and different layers can be deposited on top of each other without dissolving the underlying layer and without the need of orthogonal solvents.

Spray coating technique is suitable for industrial applications due to its ability of coating large areas at a low cost. In this technique, the solution is deposited in single droplets that are expelled by transfer gas pressure and dry immediately when hitting the surface of the substrate [6]. As mentioned above, spray coated OPDs have shown low dark current values due to the high BHJ thickness that ensures pinhole-free layers despite its high surface roughness [6]. Nevertheless, low surface roughnesses are preferred in order to improve device yield by reducing the number of spikes responsible of high dark currents and electrical short circuits.

Regarding the EQE, OPDs efficiencies are dominated by the BHJ morphology. Nanometer-sized phase separation and the existence of interconnected semi-crystalline domains between polymer and fullerene are amongst the desired characteristics [7]. Taking this into account, diketopyrrolopyrrole (DPP) electron-rich terthiophene (PDPP3T) polymer (Fig. 1a) is a good candidate for NIR OPDs. Besides its broad absorption band from 400 nm to 900 nm, it has a high molecular weight and a tendency to form semi crystalline structures in blends with PC₇₀BM, important requirements for achieving such nano-morphologies [7–14]. However, high molecular weight polymers are more difficult to dissolve and the deposition of smooth layers by spray coating can be challenging.

Therefore, the selection of the correct solvent and the use of additives become the key factor to improve BHJ morphology as well as layer topography. There is a wide list of publications that show a positive effect of 1,8-diiodooctane (DIO) on the performance of organic devices [7,9–13,15–25]. It has been shown that a small quantity of DIO added into the blend changes the crystallization of the polymer by influencing the drying process of the film. It keeps the fullerene component well dissolved in the solution allowing a better intermixing with the polymer network. As a result, an optimized nanoscale phase separation throughout the whole BHJ can be achieved [19,19,23,24]. Moreover, DIO has shown to either increase [9,23] or decrease [20] the surface roughness of PDPP3T:PC₇₀BM spin coated layers. Extensive research has been conducted to analyze the effect of DIO in PDPP3T:PC₇₀BM blends [7–15,23] for spin coated organic solar cell applications, however a deeper study in spray coated OPDs applications is still missing.

It has been shown that residual amount of DIO into the device can affect negatively the performance [26,27]. However, DIO can be removed by exposing the films to a high vacuum, a high temperature treatment or by washing out with inert solvents [26,27].

In this work, we present the main advantages of DIO in OPDs performance. Besides increasing the EQE due to the formation of better BHJ nano-morphologies, it decreases active layer roughness leading to lower and more stable dark currents at high reverse bias. It also enhances the detectivity and the linearity over five orders of magnitude and improves the crystallinity of the blend enriching charge transport and the dynamic behavior of the device.

2. Results and discussion

Fig. 1c compares the dark current performance for all devices. Apart from a reduction of the dark current at reverse bias, samples with DIO show higher dark forward currents as well as a significant increment of the EQE (Fig. 1d).

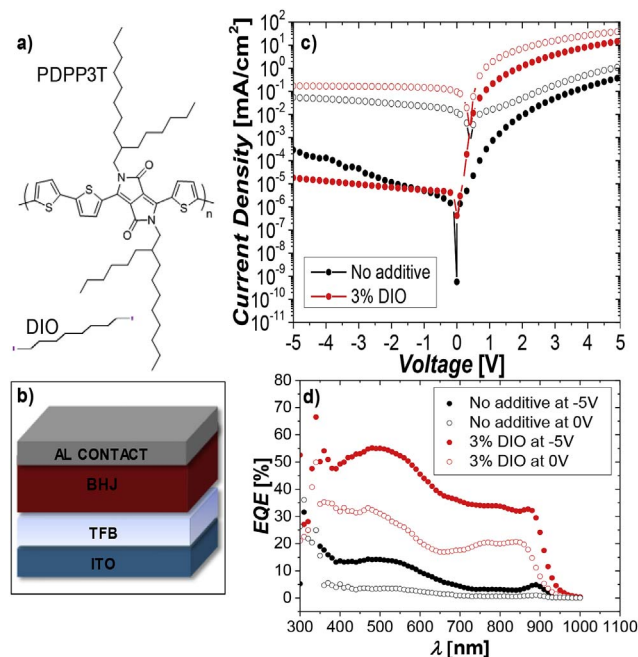


Fig. 1. a) Molecular Structure PDPP3T and DIO b) Standard structure stack of the OPD device. c) Current density-voltage (J-V) characteristics under dark (solid symbols) and under green light (open symbols) at 532 nm 780 μW/cm² for OPDs with 500 nm BHJ PDPP3T: PC₇₀BM (black) and PDPP3T: PC₇₀BM + 3% DIO (red). d) External quantum efficiency (EQE) of OPDs. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

As mentioned before, these results are based on BHJ nano-morphology improvements. Fig. 2 shows energy filtered TEM (EFTEM) images of the blends with and without DIO. Zero-loss filtered bright field TEM images (Fig. 2a and e), pre-carbon images (Fig. 2b and f) and elemental maps of sulfur (Fig. 2c and g) indicate the clear phase separation between the fullerene phases, depicted as dark contrasts that are surrounded by the bright polymer network. Elemental maps of carbon (Fig. 2d and h) confirm the corresponding phase assignment of the fullerene and the polymer network; rich phase domains correspond to the fullerene and the surrounding network to the polymer. As observed in Fig. 2e–h, DIO is reducing the domains sizes. The average particle diameter decreases from 68 nm to 31 nm. DIO is inhibiting fullerene aggregation and improving polymer networking throughout the whole BHJ. This morphology optimization improves efficient exciton dissociation at the interfaces and can be one of the reasons for higher photocurrents and EQEs. Likewise, the improved electron/hole pathways lead to charge transport enhancement which can explain the higher forward dark currents observed for samples with the additive.

The EQE values obtained for the binary solvent system (host solvent + DIO) shown in Fig. 1d are similar to the ones reported previously [10]; EQEs of 35% @ -5 V and 25% @ 0 V at 900 nm are reached. However, is important to highlight that the photo-active layer of our devices is much thicker and the amount of DIO (3%) is much lower compared to devices reported in other works. Therefore the EQE values reported here are worthy to show. Moreover, as reported by Bijleveld et al. [11] the EQE values also depend on the molecular weight of the polymer that can vary from batch to batch even for the same polymer.

Zhou et al. [12] Wu et al. [13] reported similar EQE values and Ye

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