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Effect of annealing dependent blend morphology and dielectric properties on the performance and stability of non-fullerene organic solar cells



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ABSTRACT

This work demonstrates the influence of annealing temperature on the phase morphology of the polymer-nonfullerene blend and its relationship with the dielectric constants of the blend layer. It is also the first study to report the annealing induced changes in dielectric environment of the active layer blend employing non-fullerene acceptor. Polymer solar cells (PSCs) annealed at 80 °C were found to possess optimal blend morphology favourable for efficient charge separation owing to reduced coulomb capture radius and relatively high dielectric constant, resulting in high power conversion efficiency (PCE) of \sim 11.4%. The high performing devices fabricated at optimized temperature also exhibited superior charge transport characteristics including high recombination resistance (R_{rec}) which is 31% higher than the non-annealed devices. The effect of intermolecular aggregation induced by the annealing temperature and its relationship with the disorder states in the blend layer, influencing the electron transport properties is also investigated in detail. Furthermore, a month long degradation study investigating the operational stability of the as-developed non-fullerene based PSCs was also performed and analysed.

1. Introduction

There is a great deal of worldwide interest and research aiming to develop alternative sources of energy. Photovoltaics are a common alternative source of energy with the advantages of being clean and renewable. Although silicon based solar cells are leading the PV industry right now, it is desirable to explore other photovoltaic materials that are more flexible, low-cost, light weight, transparent, suitable for rollto-roll manufacturing with short fabrication and installation time, compared to silicon and of course, the fossil fuels. Solution processed bulk heterojunction (BHJ) polymer solar cells (PSCs) meet all these requirements [1-4] and hence these are becoming appealing as an alternative energy source. Traditionally, fullerene derivatives, especially [6,6]-phenyl-C₇₁-butyric acid methyl ester (PC₇₁BM), are used in high efficiency PSCs (efficiency > 10%) as electron acceptor, along with the polymer electron donor [5-12]. Despite the dominance of fullerene acceptors in PSCs, non-fullerene acceptors are receiving considerable attention due to their excellent thermal stability [13], broad absorption spectrum [14], easily tuneable energy levels and low-cost production [15–19]. A novel non-fullerene acceptor material, 3,9-bis(2-methylene-(3-(1,1-dicyanomethylene)-indanone))-5,5,11,11-tetrakis(4-hex-

ylphenyl)-dithieno[2,3-d:2',3'-d']-s-indaceno[1,2-b:5,6-b']dithiophene

(ITIC), along with a novel donor 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), has recently broke the barrier of 11% efficiency in single junction PSCs [20]. The performance of these devices can be improved further via meticulous engineering of the donor-acceptor interfacial properties and concomitant dielectric characteristics of the material.

In particular, the influence of nanoscale morphology and related effect on the dielectric properties of non-fullerene PSCs require careful investigation. The engineering of phase morphology i.e. the mixing of the donor and acceptor (D/A) materials in the photoactive layer, is crucial for the PSC device performance due to the large exciton binding energy and small exciton diffusion length [21]. Specifically the phase morphology should be tuned in such a way that it exhibits efficient charge separation at the D/A interface owing to reduced coulomb capture radius and high dielectric constant. One way to tune the phase morphology is thermal annealing where it facilitates the temperaturedependent aggregation of donor-acceptor phases resulting in electronically favourable D/A interface [22–28]. Hence, proper optimization of thermal annealing is crucial for device performance and the optimum temperature may vary depending on the photoactive material system.

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For example, thermal annealing of P3HT:PC₆₁BM blends, at around 150 °C, creates a bicontinuous morphology with crystalline polymer domains and enhance device efficiency [24,29,30]. On the other hand, for amorphous polymers, such as PTB7, no thermal annealing is required to achieve high efficiency. The addition of additive (for example, 1, 8-diiodooctane (DIO)) helps to form small size domain that improves exciton harvesting in the fullerene and charge separation in the polymer and suggests a mixed phase is not beneficial to such device performance [31]. Though, the nanoscale phase separation processes in conventional fullerene based PSCs have been heavily studied; such indepth investigation is lacking in the case of non-fullerene PSCs and hence more investigation is required.

In this work, we present an interesting relation between the annealing temperature and phase morphology, subsequently correlating with the photovoltaic performance of polymer–non-fullerene (PBDB-T: ITIC) PSCs. The effect of annealing dependent phase morphology on the dielectric properties of the blend layer is also investigated. A systematic study of nanoscale morphology and surface topography is performed. In addition, a detailed investigation on the electronic properties and related charge recombination dynamics is done using electrochemical impedance spectroscopy (EIS). The charge transfer characteristics of annealed and non-annealed devices and its relationship with the dielectric constant are discussed in detail via EIS and Mott-Schottky analysis. We also tracked the device performance of the annealed PSCs for a month to examine the effect of annealing temperature on the device stability as well.

2. Experimental details

2.1. Device fabrication

ITO-coated glass substrates (12 mm imes 12 mm) were used for all devices. They were first cleaned by ultrasonication in soapy deionized (DI) water, DI water, acetone, and isopropanol. ZnO sol-gel solution (0.48 M) was prepared by dissolving zinc acetate dihydrate (Zn $(CH_3COO)_2$ ·2H₂O, Sigma-Aldrich, > 99.0%, 0.109 g) and ethanolamine (NH₂CH₂CH₂OH, Sigma-Aldrich, > 99.5%, 32 µL) in 2-methoxyethanol (CH₃OCH₂CH₂OH, Sigma- Aldrich, 99.8%, anhydrous, 1 mL). It was then spin-cast on top of the pre-cleaned ITO glass substrates at 4000 rpm for 60 s. These samples were then annealed at 170 °C for 30 min. PBDB-T and ITIC (purchased from Solarmer Materials Inc.) were mixed (1:1 wt ratio) in a 20 mg mL⁻¹ chlorobenzene solution with 0.5 vol% DIO in a N2-filled glovebox and stirred overnight at 50 °C. The solution was spin-coated at 2500 rpm for 1 min to obtain approx. 100 nm thick active layer. The coated substrates were loaded in a vacuum chamber at a pressure of 10^{-4} Pa, where a 10 nm film of MoO₃ and 100 nm film of silver were deposited through a shadow mask by thermal evaporation. The device area used was 0.12 cm^2 . Then, the devices were annealed at different annealing temperatures, ranging from 0 to 160 °C. At least ten devices were made for each case.

2.2. Device characterization

The current density-voltage (J-V) measurements were performed using a solar cell I-V testing system from PV Measurements, Inc. (using a Keithley 2400 source meter) under illumination power of $100 \ \text{mW}/$ cm² by an AM 1.5G solar simulator. The devices were stored in a N₂filled glovebox prior to J-V characterization in air within 2 days of fabrication to minimize the effects of device degradation on results. During characterization, the temperature and humidity of the characterization area were controlled by the use of forced air flow and dehumidifier (maker: Ausclimate, model no: WDH-930DA); respectively, to minimize thermal and moisture induced degradation effects. The device temperatures (maintained at 25–30 °C) were measured by GM1350 50:1 LCD Infrared Thermometer Digital Gun. The relative humidity of the test area was \sim 35%. However, the devices retained the initial performance for at least 1 month after fabrication when stored in the glovebox under dark conditions at room temperature. External quantum efficiency (EQE) measurements were performed using a QEX10 spectral response system from PV measurements, Inc. For optical characterization, a UV-VIS-NIR spectrometer (Perkin Elmer -Lambda 950) was used. The film surface morphology was observed by atomic force microscopy (AFM) with a Bruker Dimension ICON SPM (scan size: $5 \mu m \times 5 \mu m$; scan rate: 0.407 Hz; samples/line: 512). Surface topology image was captured by FEI Nova NanoSEM 230 FE-SEM. The EIS and capacitance-voltage (C-V) measurements were performed in a N2-filled glovebox with an Autolab PGSTAT-30 equipped with a frequency analyser module in the frequency range of 10^{6} –100 Hz. AC oscillating amplitude was as low as 20 mV (rms) to maintain the linearity of the response. Film thickness was estimated by a KLA Tencor D-600 Stylus Profiler with sub-Angstrom resolution.

3. Results and discussions

We fabricated PSCs with device structures of ITO (Indium tin oxide)/ZnO/PBDB-T:ITIC/MoO₃/Ag, where the ZnO and MoO₃ were used to fabricate the n -type and p -type charge transport layers, respectively (Fig. 1(a)). The chemical structures of the polymer donor (PBDB-T) and non-fullerene acceptor (ITIC) are shown in Fig. 1(b). The fabricated devices were annealed at different temperatures within a broad range (0–160 °C) to find the optimum annealing condition. To determine the best-performing PSCs, the current-density-voltage (J–V) curves measured under simulated AM 1.5G, 100 mW/cm² irradiance are shown in Fig. 2(a) and the corresponding photovoltaic parameters



Fig. 1. (a) Schematic diagram of device structure of the fabricated inverted PBDB-T:ITIC OPV devices; (b) chemical structures of active layer materials (donor polymer: PBDB-T and non-fullerene acceptor: ITIC).

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