



Controlling intercalations of PBDTTT-EFT side chain to initiate suitable network for charge extraction in PBDTTT-EFT:PC₇₁BM blended bulk heterojunction solar cell



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ABSTRACT

Electrical, optical and structural properties, based on blend of PBDTTT-EFT:PC₇₁BM bulk heterojunction solar cell is investigated. We have shown the presence of PC₇₁BM in the blend facilitates interfaces for charge transfer mechanisms leading to PCEs of ~ 9.38%. GIXRD line profile obtained reveals that quantitative presence of PC₇₁BM initiates a change in preferred orientation. It is proposed that PC₇₁BM facilitates polymer alkyl side chain intercalations causing reduced d_{100} spacing, leading to larger π - π stacking of the back bone. The data obtained from d_{200} plane associated with PC₇₁BM, reveals correlation between increased vertical stacking and PC₇₁BM loading, suggesting a bicontinuous network for electron transport. Raman spectroscopy results provides evidence of PC₇₁BM impact on frequency shift associated with C = C stretching mode of side chain thiophene rings responsible for of π - π spacing. Correlations between AFM surface roughness and blend ratio reveals migration of PC₇₁BM to the film surface.

1. Introduction

Organic solar cells have been the subject of extensive investigation for more than three decades owing to properties such as: high optical absorption coefficient, ultrafast electron transfer, solubility, flexible substrate, room temperature and lower fabrication costs [1–4]. Whilst OPV devices manufactured from blend of acceptors and relatively large band gap polymers were identified as promising active material for future generation of solar cells [5,6], nonetheless, substantially lower associated PCEs are disappointing when compared to its inorganic counterparts. Several communications associated with the impact of thermal and vapour annealing [7–11], buffer materials [12–14], Nano-structuring [15,16] is reported to incrementally increase PCEs but not substantially to manifest in breakthrough in solar cell manufacturing. Recent advances in material synthesis paved the way for realisation of narrow band gap polymers such as PBDTTT-EFT, PTB7, PBDTTT-CT and PBDTTT-CF, resulting in PCEs of 7–9% [17–20]. To improve PCE of narrow band gap devices, researchers focused their attention on processes applied for enhancing performance of large band gap devices [21–23]. Whilst some processes such as thermal annealing of large band gap material had positive impact on PCEs, it was proved to be detrimental in the case of narrow band gap polymer PBDTTT-EFT [24–26]. The presence of different acceptors in blended solution with large band

gap polymers proved to be crucial in open circuit voltage value and exciton dissociation leading to higher PCEs [27]. Recently the impact of PC₇₁BM percentage loading on narrow band gap polymer PBDTTT-EFT was investigated, giving supporting evidence that, fullerene concentration regime plays an important role in exciton dissociation and charge transport [28,29]. We propose that the role of PC₇₁BM and its quantitative presence in blended solution with PBDTTT-EFT is multi-purpose processes which some results in improved PCEs whilst, some having opposing effects. To this end the role of PC₇₁BM and its quantitative presence in blended solution with PBDTTT-EFT is investigated to elucidate the Nano-structural formation and its subsequent impact on thin films electrical behaviour.

2. Material and methods

Pre-fabricated ITO/glass substrates were first cleaned using ultra sonic bath with three step process; deionised water, acetone, iso-propanol and left to dry in nitrogen environment. PEDOT: PSS purchased from Ossila was filtered and then spin coated onto the substrate at a speed of 5000 rpm for 30 s and then baked on a hotplate at 120°C for 10 min. A DektakXT thickness profiler accurately measured the film to be 30 – 40 nm. The substrate was then transferred to a nitrogen glove box maintained at 0.1 ppm for O₂ and H₂O level left for 30 min before

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fabricating the active layer. PBDTTT-EFT: PC₇₁BM (1-Material, used as received) with concentration of 25 mg mL⁻¹ in 1,2-dichlorobenzene but with blend ratios of 1:0.5, 1:1, 1:1.15, 1:2 and 1:3 were produced. The pre-tested PBDTTT-EFT: PC₇₁BM spin-casted at 600 rpm for 18 s resulted in a film thickness of ~ 100 nm. After fabrication of the active layer, substrates then vacuum dried for 5 min at -100 kPa ready for surface washing with 60 μL of Ethanol purchased from Sigma Aldrich and drop cast at 4000 rpm for 30 s. For fabrication of top electrodes, substrate was transferred to a metallisation rig (Auto 500) via an interconnecting chamber. First 10 nm of Calcium (sigma Aldrich) was fabricated at a rate of 0.1 nm s⁻¹ using mask aligner. After a delay time of 5 min 100 nm of Aluminium was then deposited through the mask aligner at a rate of 0.1 nm s⁻¹ for the first 20 nm and then at a rate of 0.5 nm s⁻¹ for the remaining 80 nm. A microbalance quartz crystal monitor (Intellmetrics IL 150) was used to measure the deposition rate as well as the Aluminium thickness. During the deposition, the chamber vacuum was maintained below 10⁻⁶ Torr. For Raman, PL and GIXRD studies similar processes were followed apart from metallisation stage.

The active area for each device was 0.13 cm², illuminated through a shadow mask under 1 SUN condition using 1.5 AMG filter (LOT-LSZ389) and xenon arc lamp solar simulator (LOT-LS0306). I-V characteristics were collected using Keithley 2400 source meter. To test the accuracy of solar simulator, silicon reference solar cell (LOT-LS0041) which its accuracy is certified by National renewable energy laboratory was used to adjust the input power.

The GIXRD data were collected using Bruker D8 Advance in out-of-plan XYZ stage mode. Optical absorption data were collected using (Varian Cary 50-Bio). Raman and PL measurements were collected using (Renishaw inVia) with 685, 785 nm excitation laser. The device morphology was performed using Agilent AFM 5400 series.

3. Results and discussion

3.1. Optical and electrical characterisations

PBDTTT-EFT: PC₇₁BM blend with ratios 1:0.5, 1:1, 1:1.15, 1:2 and 1:3 were fabricated either for optical and structural investigations or electrical characterisations. PBDTTT-EFT is a narrow band gap copolymer used as donor material in our devices structure [30,31]. The optical absorption spectrum and the impact of PC₇₁BM in the blend is fundamentally important to this investigation. For this reason, optical absorption spectroscopy based on blended films with increasing PC₇₁BM loading is investigated and the results are presented in Fig. 1a. As is evident upon introduction of PC₇₁BM and subsequent increase in its ratio, a redshift in the peak associated with π-π stacking of PBDTTT-EFT backbone unit is observed [32]. Furthermore, the onset of optical absorption is found to shift towards the longer wavelength with increasing PC₇₁BM blend ratio. This could initiate a change in optical band gap and result in variation of open circuit voltage as suggested in the literature [29]. To verify the impact of PC₇₁BM on open circuit voltage, electrical characterisations were performed and graphically shown in Fig. 1b. Results presented in Table 1, indicate that the power conversion efficiency, peaks at blend ratio of 1:2 with further increase leading to the deterioration of this parameter. An interesting observation made is that increasing PC₇₁BM loading has no impact on open circuit voltage in contrast to the increase of this parameter previously reported [29]. This is explained to be the result of PC₇₁BM aggregation when its percentage loading is increased, initiating reduction of tail state in the density of states (DOSs) leading to higher V_{OC} [29]. We believe this may be the result of solvent used and film drying process facilitating PCB₇₁M aggregation. Furthermore, the short circuit current density peaks at 1:1.5 blend ratio but relatively lower value of fill factor results in PCEs of approximately 8.08%. However, this parameter further improves to 9.38% upon increasing PC₇₁BM loading to 1:2 at the expense of improved fill factor even though a reduction in the short circuit current density is observed. To gain insight to the trend of electrical parameters

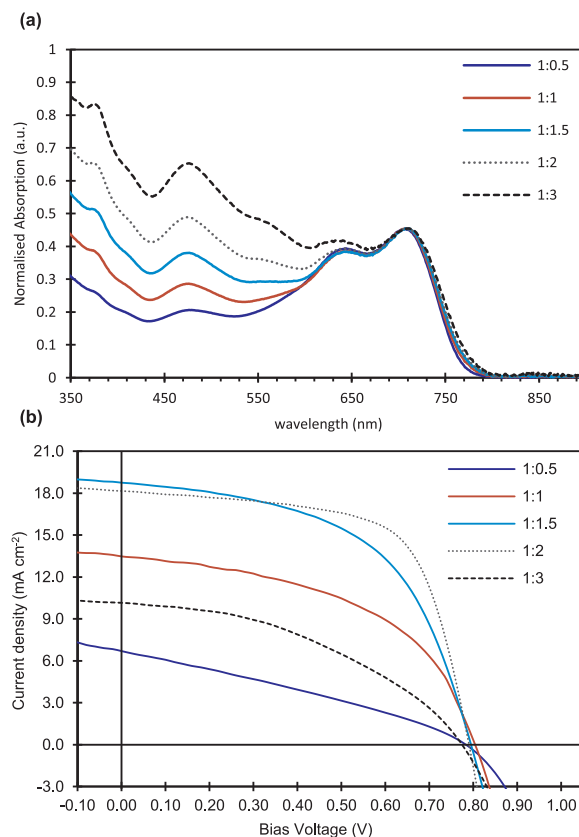


Fig. 1. a) Optical absorption spectrum for PBDTTT-EFT: PC₇₁BM blend ratios, b) J-V characteristics for PBDTTT-EFT: PC₇₁BM blend ratios.

presented in Table 1 and to elucidate the trade-off between short circuit current density and FF for 1:1.5 and 1:2 PC₇₁BM loading, photoluminescence measurements were performed on thin films devices with the same blend ratios. As shown in Fig. 2a the PL intensity quenches by introducing PC₇₁BM resulting from a fast charge transfer [9,33–35]. Upon further PC₇₁BM loading the quenching efficiency improves as shown in Fig. 2b and peaks at 1:1.5 ratio which corresponds to the highest short circuit current density observed. Similar observations indicate that the best quenching is obtained at 1:1.5 PBDTTT-EFT:PC₇₁BM ratios [28]. We believe 1:1.5 blend ratio provides domains suitable for efficient exciton dissociation resulting in enhanced short circuit current density. However, further increase in PC₇₁BM above 1:1.5 leads to higher PL intensity yielding lower short circuit current density [28]. We suggest increasing PC₇₁BM above 1:1.5 ratio could facilitate opposing processes initiating lower exciton dissociations at the same time as providing suitable networks for charge extractions. Fig. 3 shows correlation between the device fill factor and shunt resistance as a function of blend ratio. As is evident, both these parameters peak at 1:2 ratio as presented in Table 1. We believe improved value of fill factor is due to better charge carriers transport network and lower device leakage current. Previous work by Foster et-al [36] suggests that the improved value of fill factor is initiated from increased PC₇₁BM loading, providing suitable network and leading to higher electron mobility. Further work on other polymers also suggests similar behaviour upon PC₇₁BM loading [37].

Fig. 4a reveals the AFM topography and phase images of PBDTTT-EFT:PC₇₁BM blended films. When AFM surface roughness (Sq) is measured, it appears that there is correlation between this parameter and series resistance presented in Table 1 and Fig. 4b. As can be observed the lowest series resistance corresponds to lowest value of Sq. We believe this stems from better surface contact, resulting in lowering R_s. Similar trend has been reported for other polymers/fullerene blended

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