

Evolution of phase separation upon annealing and the influence on photocurrent generation in ternary blend organic solar cells



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ABSTRACT

The morphological changes by thermal annealing and their impact on photocurrent generation in binary and ternary blend organic solar cells were studied with photoluminescence measurements. Specifically the influence of adding the polyfluorene copolymer poly(9,9-dioctylfluorene)-2,7-diyl-alt-[4,7-bis(3-hexylthiophen-5-yl)2,1,3-benzothiadiazole]-2',2''-diyl (F8TBT) to poly(3-hexylthiophene) (P3HT):[6,6]-phenyl-C₆₁-butyric acid methyl ester (PCBM) blends was investigated. This third constituent (F8TBT) has ambipolar charge transport properties and acts as an efficient electron donor when paired with PCBM and electron acceptor when paired with P3HT. While in principle the addition of a third component may assist charge separation via an energy cascade-type process, the photovoltaic efficiency of ternary blends was found to be inferior to that achieved with binary blends. We discuss reasons for this lower efficiency and what factors need to be considered for the third component to improve the efficiency of organic ternary blend solar cells.

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1. Introduction

Organic solar cells require a combination of donor and acceptor components to dissociate the tightly bound excitons formed by photoexcitation in organic semiconductors [1–5]. In solution-processed devices these components are often combined in a bulk heterojunction structure where the donor/acceptor interface is distributed throughout the absorbing layer [6–9]. Even once a charge-transfer state has been produced at the donor/acceptor interface, significant Coulombic binding between the electron and hole is expected, and there is still considerable debate about how efficient charge separation (as opposed to geminate recombination) is achieved in the best cells [10–15]. One strategy to prevent recombination is to arrange a “cascade” structure where electron transfer from the photoexcited donor, for example, proceeds via a thin intermediate layer, followed by a further downhill electron transfer event onto the final acceptor material [16–19]. This strategy, as used by photosynthesis, provides rapid spatial separation of the electron and hole, thus suppressing recombination. Cascade structures have been investigated with some success in planar device geometries, but are hard to achieve in bulk heterojunction devices where we do not have access to the interface [20]. Recent evidence

suggests that cascade energy landscapes may occur naturally in certain polymer:fullerene blends, where a mixed polymer:fullerene phase acts as the intermediate layer between purer polymer and fullerene phases [17,19]. In principle, it might be possible to form such a structure by forming a ternary blend of the three components, although in practice it is difficult to optimize the morphology of such blends [20–25].

In this paper, we study the effect of adding the polyfluorene co-polymer poly(9,9-dioctylfluorene)-2,7-diyl-alt-[4,7-bis(3-hexylthiophen-5-yl)2,1,3-benzothiadiazole]-2',2''-diyl (F8TBT) to blends of the well-studied poly(3-hexylthiophene) (P3HT): [6,6]-phenyl-C₆₁-butyric acid methyl ester (PCBM) system. Since it has been shown that F8TBT can act as an efficient electron donor in combination with PCBM, and an efficient electron acceptor when paired with P3HT [26,27], there is the possibility of enhanced charge separation through sequential charge transfer first from P3HT to F8TBT and then F8TBT to PCBM. However we find that the efficiencies of P3HT:F8TBT:PCBM ternary blends were inferior to those of P3HT:PCBM bulk heterojunction solar cells, and investigate the reasons why the addition of F8TBT results in poorer device performance. Such a study is important for understanding the criteria required for the success of such ternary blends.

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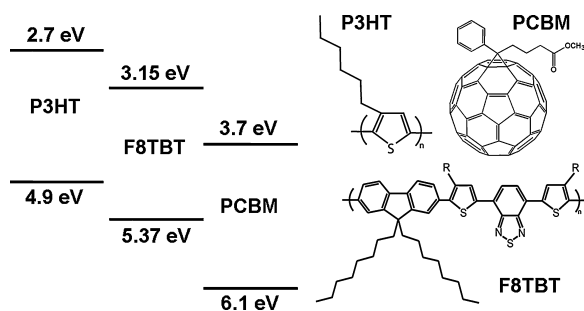


Fig. 1. The chemical structure and the energy diagram of P3HT, PCBM and F8TBT.

2. Experimental methods

The chemical structure and the energy diagram of P3HT, F8TBT and PCBM are illustrated in Fig. 1. P3HT and F8TBT were provided by Rieke Metals and Cambridge Display Technology Ltd, respectively. PCBM was purchased from Nano-C (99.5% purity). The weight ratio of P3HT and PCBM in chlorobenzene solution was 1:0.8. F8TBT was systematically added to the P3HT:PCBM blend up to a 1:1 weight ratio with respect to PCBM with an increment of 20% P3HT weight: The weight ratios of P3HT:F8TBT:PCBM are 1:0:0.8, 1:0.2:0.8, 1:0.4:0.8, 1:0.6:0.8 and 1:0.8:0.8. Quartz substrates and glass substrates covered with an ITO strip were used for films and device fabrication, respectively. All the substrates were cleaned by sonication in acetone and isopropanol for 10 min each, and then were plasma-etched in oxygen plasma at forward 250 W and reverse 0 W for 10 min. For devices, PEDOT:PSS filtered with a hydrophilic filter was spin coated on glass/ITO substrates with 6000 rpm for 60 s, resulting in 40 nm thickness. Samples with PEDOT:PSS were annealed at 150 °C for 30 min under nitrogen purge, and then immediately transferred to the glovebox. The solutions in chlorobenzene prepared in the glovebox were spin coated on quartz substrates and the PEDOT:PSS substrates. For device fabrication, prior to annealing samples at various temperatures for 10 min on a hot plate, aluminum with approximately 100 nm thickness was deposited on the samples by thermal evaporation at roughly 10^{-6} mbar. As electrical characterization was carried out in air, devices were encapsulated with epoxy and cover glasses to avoid oxygen and water contamination.

External quantum efficiency (EQE) was measured as a function of wavelength by dispersing light from a tungsten filament lamp through a monochromator with a spot size no larger than the device active area. Light intensities of less than 0.1 mW cm^{-2} were used with short-circuit current recorded using a Keithley 237 source measure unit. Current–voltage characteristics were acquired under simulated sunlight (100 mW/cm^2 AM1.5G global spectrum) from an Oriel 81160-1000 solar simulator calibrated to a silicon reference cell.

Absorbance spectra were collected with Hewlett Packard UV–vis spectrometer. Photoluminescence spectra were recorded with a Varian Cary Eclipse integrated fluorescence spectrometer by exciting the samples at 500 nm. Photoluminescence quantum efficiencies (PLQY) were determined as described by de Mello et al. [28].

Atomic force microscopy (AFM) images are not shown as the morphology shown in the AFM images was too fine to analyze phase separation.

3. Results and discussion

Fig. 2 presents the absorption and the PL spectra of neat P3HT, F8TBT, PCBM, and their as-spun binary blend films with 1:1 weight ratio. P3HT has reasonably distinct vibronic structure due to its

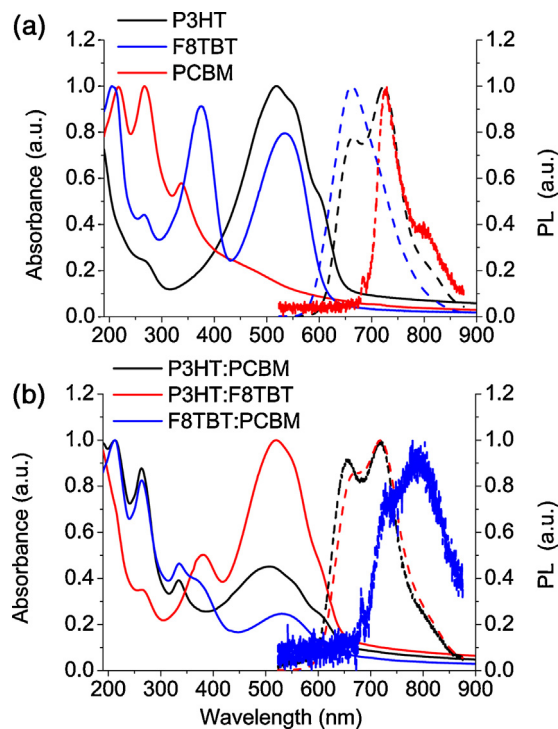


Fig. 2. Absorption and PL spectra of (a) pristine films and (b) binary blend films. All spectra are normalized to the maximum.

semicrystalline structure in film [29]. The absorption band of P3HT overlaps the lower absorption band of F8TBT, indicating that the energy gap of the two is almost identical. PCBM has a wider band gap, as shown in the absorption spectrum. PL spectra were obtained with excitation at 500 nm, where both P3HT and F8TBT absorb light.

To explicitly investigate energy and charge transfer processes, the PLQYs of as-spun films made of P3HT:F8TBT, F8TBT:PCBM, P3HT:PCBM binary blends were measured and compared to the PLQYs of neat films. P3HT has PLQY of ~5%; F8TBT is the most fluorescent among the three, ~35%; and PCBM is very weakly fluorescent such that the PLQY could not be accurately determined. For P3HT:PCBM and F8TBT:PCBM blends the PLQYs are too low to be determined, but P3HT:F8TBT binary blends have PLQYs of ~3%. The emission of the F8TBT:PCBM film is red-shifted compared to both F8TBT and PCBM, suggesting that the emission arises from charge-transfer complexes forming at the interfaces of F8TBT and PCBM. Although P3HT:F8TBT is fluorescent relative to the other binary blends, all the binary blends are shown to quench excitons before their radiative recombination, which indicates that charge transfer efficiently occurs.

It is well known that P3HT reorganization occurs upon thermal annealing, enhancing photovoltaic performance [30–34]. In order to find out the optimum fabrication condition for the ternary blend solar cells with respect to thermal treatment, we first found the optimum thermal conditions for P3HT:PCBM blend solar cells. The optimum annealing temperature was 110 °C. The EQE spectra at different annealing temperatures between 90 and 130 °C are almost indistinguishable (data now shown), but the power conversion efficiency shows that P3HT:PCBM has the highest efficiency of 3.1% when annealed at 110 °C after aluminum deposition by thermal evaporation on the active layer (cf. Table 1).

We note that the shapes of the EQE spectra among the binary blend solar cells are different: the P3HT:PCBM binary blend solar cell has a peak EQE at 500 nm, P3HT:F8TBT a peak EQE at 550 nm and F8TBT:PCBM a peak EQE at ~480 nm (see Fig. 3) [26]. An EQE spectrum is in general correlated with the spectrum of the

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