

Comparative study of spectral and morphological properties of blends of P3HT with PCBM and ICBA

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ABSTRACT

We report a comparative study on spectral and morphological properties of two blend systems for polymer solar cells: the donor material poly(3-hexylthiophene) (P3HT) in combination with the acceptor material of either [6,6]-phenyl-C₆₁ butyric acid methyl ester (PCBM) or indene-C₆₀ bisadduct (ICBA) that was reported to enhance efficiencies of polymer solar cells. Optical microscopy and grazing incidence X-ray scattering reveal the stronger tendency of PCBM to form larger and more ordered domains/grains than ICBA either in pure or blend films. Compared to PCBM, the presence of ICBA also substantially perturbs the organization and longer-range ordering of P3HT in increasing the ICBA ratio in blends. With larger and more ordered phase-separated domains, the P3HT/PCBM blend films exhibit significant optical scattering at higher PCBM ratios. Yet, such optical scattering is not significant for P3HT/ICBA blends (even with high ICBA ratios). Overall, results here suggest the reported higher efficiencies of P3HT/ICBA solar cells (vs. P3HT/PCBM cells) cannot be attributed to larger and/or more ordered phase-separated donor-acceptor domains and other characteristics play more important roles in this case.

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1. Introduction

Organic photovoltaics (OPVs) have attracted wide attention in recent years due to their potential advantages in fabrication, cost, and mechanical flexibility [1–3]. The energy conversion process in OPVs typically involves exciton generation upon light absorption, exciton diffusion and then dissociation of excitons into free carriers at the donor–acceptor (D–A) interface, and finally transport of carriers to their respective electrodes [1–3]. To achieve high conversion efficiencies, polymer bulk-heterojunctions (BHJ) composed of nanoscale interpenetrated (phase-separated) domains of donor and acceptor materials are

introduced to increase areas of D–A interfaces for ensuring effective exciton diffusion to such interfaces, meanwhile providing transport paths for carrier transport/extraction to electrodes [4]. Among various D–A materials, poly(3-hexylthiophene) (P3HT) as a donor and [6,6]-phenyl-C₆₁ butyric acid methyl ester (PCBM) as an acceptor are the most widely studied D–A combination [5–14], in considering light harvesting, carrier generation and carrier transport. Regioregular P3HT are known to have strong tendency of forming self-organized structure in thin films, which could be further enhanced upon appropriate treatments (e.g. thermal annealing, solvent annealing, etc.) [6–12]. More ordered molecular packing benefits charge transport (i.e., higher mobility) and absorption at longer wavelengths due to enhanced inter-chain interactions [6]. In BHJs, such treatments induce not only re-crystallization but also inter-diffusion of D/A

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components, giving some flexibility in tuning/optimizing nanoscale phase separation and interpenetration of donors and acceptors that is essential and critical to high-efficiency BHJ OPVs [13,14].

Recently, a few reports show that replacing PCBM with a new C_{60} -based acceptor material indene- C_{60} bisadduct (ICBA) in the P3HT/fullerene system could substantially improve the conversion efficiency of the solar cells [15–18]. Compared to PCBM, ICBA has more facile synthesis, higher solubility in common organic solvents, and stronger visible absorption for better light harvesting in the photovoltaic process. ICBA also exhibits a higher LUMO level than PCBM [15–18], which contributes to the higher open-circuit voltage of the P3HT/ICBA photovoltaic cell. In addition to the above properties, the detailed morphological properties of the donor–acceptor composites would also be essential in understanding the difference in performance between the P3HT/PCBM system and the relatively new P3HT/ICBA system. Although there have been numerous studies of the morphological properties of the P3HT/PCBM system, yet thus far the morphological study on the P3HT/ICBA system is sparse [19]. To acquire better understanding and useful information, in this work, a comparative study on morphological and related properties of the P3HT/PCBM and the P3HT/ICBA systems is conducted.

2. Experiments

2.1. Preparation of polymer/fullerene films

The structures of P3HT, PCBM, and ICBA used in this study are shown in Fig. 1. Regioregular P3HT (average molecular weight = 55–60 K, regioregularity = 95%, PDI ~ 2.0), PCBM, ICBA were purchased from Rieke Metals Inc., Nano-C, and Luminescence Technology Corp., respectively, and were used as received. The pure or blend P3HT/fullerene solutions having different P3HT and fullerene weight ratios [P3HT: fullerene (PCBM or ICBA) = 1:0, 3:1, 2:1, 1:1, 1:2, 1:3, 0:1] were all prepared using chlorobenzene as the solvent and a total weight concentration of 20 mg/mL. The solutions were thoroughly stirred and shaken at the room temperature before use for film coating. Pure or blend thin-film samples of P3HT/fullerene for various studies were prepared by spin-coating (at ~700 rpm) on Corning E2K glasses, with the average film thickness of

~120 nm. For samples subjected to annealing, the annealing was conducted at 150 °C (the annealing temperature usually used for BHJ polymer solar cells) for 20 min.

2.2. Characterization of polymer/fullerene films

A UV–vis spectrophotometer (V-670, JASCO) equipped with an integrating sphere was used to characterize the transmittance, reflection and absorption spectra of pure or blend P3HT/fullerene films. Since some thin-film samples of P3HT/fullerene blends showed optical scattering properties, in this work, two types of transmittance were characterized: the total transmittance (T_{total}) and the direct transmittance (T_{direct}). T_{total} was measured by using a monochromatic light beam normally incident onto sample and then using an integrating sphere to collect transmitted light over all angles. On the other hand, T_{direct} was measured by using a monochromatic light beam normally incident onto the sample and then collecting transmitted light only in the normal direction (within a 5° collection angle). The difference between T_{total} and T_{direct} reveals whether the film is optically scattering. Similarly, two types of reflectance were characterized: the total reflectance (R_{total}) collected with the integrating sphere and the direct reflectance (R_{direct}) collected within a 5° collection angle. Since some thin-film samples of P3HT/fullerene blends showed optical scattering properties, the determination of optical absorption properties of films requires special attention. Instead of determining the actual absorbance (A) of various films from the normal transmittance directly (i.e., $A = 1 - T_{\text{direct}}$), the absorbance A was determined by first measuring T_{total} and R_{total} and then by $A = 1 - T_{\text{total}} - R_{\text{total}}$.

The morphologies and nanostructures (e.g. intermolecular structures and orientations etc.) of various pure and blend P3HT/fullerene films were investigated by grazing incidence X-ray scattering (GIXS). Compared to other conventional techniques of morphological characterizations (e.g. atomic force microscopy-AFM, scanning or transmission electron microscopy-SEM/TEM etc.), GIXS has the particular advantage of being able to provide structural/morphological information of a thin film at different scales [20–27], instead of being limited to just local observation or sample preparation (e.g. in AFM, SEM, TEM etc.). Fig. 2 illustrates the configuration of the GIXS measurement, which was conducted at the BL17A end station of the

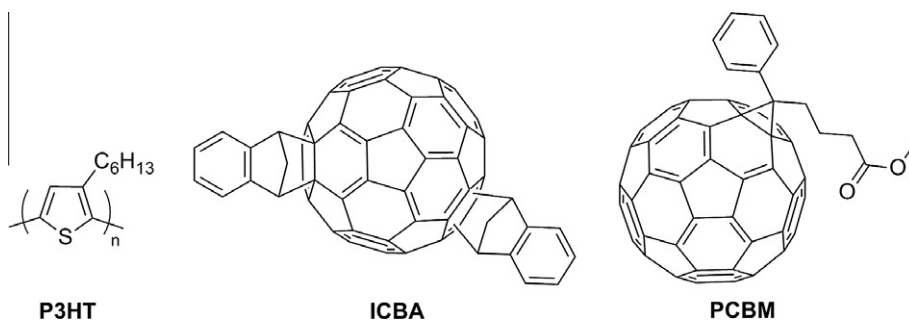


Fig. 1. Molecular structures of P3HT, ICBA, and PCBM.

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