



Efficiency enhancement of ternary blend organic photovoltaic cells with molecular glasses as guest acceptors



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ABSTRACT

Previously reported glass-forming perylene diimide (PDI-glass) and diketopyrrolopyrrole (DPP-glass) acceptors were used as ternary components to modulate the morphology and electron transport properties of bulk heterojunctions composed of the polymer blend poly(3-hexylthiophene) (P3HT) and [6,6]-phenyl-C₆₁-butyric acid methyl ester (PCBM). Both ternary acceptors show strong absorption in the visible range and energy levels intermediate between that of P3HT and PCBM. In the blends, both amorphous materials occupy the interface between P3HT and PCBM domains without significantly altering film morphology. The incorporation of 20% PDI-glass and 15% DPP-glass in ternary blend devices has resulted in conversion efficiency enhancements of 38% and 36%, respectively. Additionally, the incident photon to electron conversion efficiency of the ternary devices of these materials was increased with additional photocurrent between 660 and 750 nm, indicating that these materials efficiently contribute to light harvesting. This justifies the efficiency enhancements via the charge transfer mechanism. Furthermore, these ternary devices show high stability towards both moisture and oxygen.

1. Introduction

In recent years, organic photovoltaic (OPV) cells have attracted increased attention as potential sources of renewable energy over traditional inorganic solar cells due to their simplicity, flexibility, light weight, cost effectiveness, and processability [1–7]. However, the commercialization of organic PV cells is limited due to their relatively low power conversion efficiency and low stability as compared to silicon-based solar cells [8–10]. Traditionally, common materials used in OPV cells are poly(3-hexylthiophene) (P3HT) and [6,6]-phenyl-C₆₁-butyric acid methyl ester (thereafter referred to as PCBM). Although both materials are still considered benchmarks, various materials with significantly higher performances have been developed in recent years [11,12], though often at a significant increase in cost. P3HT therefore retains certain advantages over other donor polymers such as low cost, high crystallinity, high hole mobility, and ease of synthesis. For these reasons, while P3HT:PCBM bulk heterojunction OPV cells can reach conversion efficiencies around 5% [13], attempting to improve the performance of OPV cells using P3HT is still the object of research. However, P3HT only absorbs photons in the blue and green parts of the spectrum with wavelengths below 650 nm, leading to low short-circuit

current and low PCE [14].

The two principal limitations of OPV cells consist in their narrow absorption range, which leads to a significant portion of the visible and near-infrared spectrum not being harvested, and the control over the morphology of the bulk heterojunction, which is often suboptimal and leads to various defects that prevent the active layers from operating at their full potential. Besides designing alternative donor and acceptor materials with improved performance, several other approaches were employed to enhance the efficiency of organic photovoltaic cells, including solvent additives, solvent vapor treatment, the incorporation of metal nanoparticles in the active layer, and improving morphology for better charge transport [2,15–17]. The fabrication of tandem OPV cells by stacking two or more subcells with different donor or acceptor materials has also been attempted for enhancing the light absorption for OPV cells. However, this approach is limited because of its complexity [18,19]. Therefore, it is necessary to investigate a simpler and effective method to enhance the efficiency of OPV cells.

Ternary blend OPV cells constitute a simple alternative approach that can also expand the absorption spectra by incorporating another organic material as guest sensitizer [20]. Generally, ternary blend OPV cells can contain an active layer with either two donors and one

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acceptor, or two acceptors and one donor. In both cases, they are believed to follow the same mechanisms: the energy transfer, charge transfer, parallel-linkage, and alloy models [19,21–23]. Various types of materials such as nanoparticles, low bandgap polymers and small molecules can be incorporated as sensitizers, which typically show a complementary absorption spectrum to the polymer-based blend. Small molecules are the most promising sensitizers for ternary blends because they allow the tuning of both the absorption and the morphology of the active layer by modifying their molecular structure [24].

There are several examples reported in the literature of ternary blend OPV cells, both based on two donors and one acceptor, or with one donor and two acceptors [24–26]. Since fullerene derivatives are often used as acceptors, and show very weak absorption in the visible range, the ternary blend approach becomes especially appealing in these cases because the additional light harvesting by the third component can increase cell performance significantly. In one example, an efficiency enhancement of 20% was reported by incorporating an additional non-fullerene acceptor with strong absorption in both the visible and near-infrared ranges in the P3HT:PCBM blend [25], while a recent study using a fullerene derivative as ternary acceptor in a PBDB-T:IT-M blend has shown a 13% enhancement to reach a maximal conversion efficiency of 12.2% [26]. However, the crystalline nature of such additional components can disrupt the morphology of the blend, either upon deposition of the blend or upon prolonged operation, thereby causing a loss of efficiency [25].

Amorphous materials constitute an appealing alternative because they conveniently avoid most pitfalls associated with crystallinity [16]. Although most small molecules inevitably converge towards the thermodynamically favored crystalline state, there exists a small group of small-molecule compounds, called molecular glasses or amorphous molecular materials, that are capable of remaining amorphous for indefinite periods of time [27]. Molecular glasses are appealing because they are suitable materials for forming high-quality thin films, while retaining the advantages related to monodisperse species [28,29]. Both our group [30–32] and other groups [33–35] have demonstrated that molecular glasses could be used successfully in organic photovoltaics with both donors and acceptors, and by blending both homogeneously as an amorphous active layer. Additionally, molecular glasses were shown to be suitable as an interfacial layer in perovskite PV cells, leading to an enhancement of 39% by using a glass-forming perylene diimide (PDI) derivative [36]. In the context of ternary blend OPV cells, glass-forming materials are especially appealing as ternary components, because they can be expected to locate at the interface between the donor and acceptor domains without perturbing their respective crystalline morphology, thereby providing more pathways to extract charges while preserving the supramolecular architecture of the active layer.

Herein, molecular glasses were used for the first time in ternary blend OPV cells based on P3HT/PCBM. Inverted devices were fabricated by incorporating variable concentrations of either PDI-glass or DPP-glass, leading to enhancements reaching up to 35–38%. Interestingly, while DPP-glass shows significantly higher efficiencies than PDI-glass when used as acceptor, their impact as ternary components were closely similar, hinting that the performance enhancements observed are due to an improvement of the cohesion between P3HT and PCBM crystalline domains, coupled with an increased light harvesting. The devices showed high stability towards moisture or oxygen, with little performance degradation observed after 40 days under ambient conditions.

2. Experimental

2.1. Materials

The patterned Indium tin oxide (ITO) coated glasses used as substrates were purchased from Luminescence Technology Corporation,

with ITO film thickness approximately 135 ± 15 nm and sheet resistance $15 \Omega \text{sq}^{-2}$. Regioregular poly (3-hexylthiophene) (P3HT) used as electron donor was purchased from Rieke Metals. Molybdenum oxide used as hole transport layer and [6,6]-phenyl-C₆₁-butyric acid methyl ester (PC₆₁BM, thereafter referred to as PCBM) as electron transport layer were purchased from Alfa Aesar. Zinc oxide (ZnO) used as electron transporting material was synthesized by the sol-gel method as reported in the literature [31]. PDI-glass and DPP-glass were synthesized according to previously published procedures [30,31].

2.2. Device fabrication

Devices were fabricated according to a previously published procedure [37, 38]. Pre-patterned ITO substrates obtained from commercial sources were cleaned consecutively in an ultrasonic bath with soapy water, distilled water, acetone and isopropyl alcohol for 15 min each and finally dried with air. The substrates were heated on a hot plate at 80–90 °C for 20 min to improve the surface adhesion and wettability of ITO-glass [39]. The dried substrates were further cleaned with a plasma cleaner for 15 min. Zinc oxide (ZnO) precursor gel was spin-coated on cleaned ITO substrates at a rate of 3000 rpm in air. The substrates coated with ZnO were baked at 160 °C for 1 h. The thickness of zinc oxide was approximately 25–30 nm as measured with a Sloan Dektak II profilometer. Blend solutions of P3HT and PCBM (total concentration 35 mg/mL) were prepared in dichlorobenzene in 1:0.8 wt ratio and stirred for 24 h at 50 °C. Various weight ratios of PDI-glass or DPP-glass were added to P3HT: PCBM blend solutions and stirred 2 h before spin-coating. The solutions were filtered through a 0.45 μm poly (tetrafluoroethylene) (PTFE) filter before active layer deposition. The active layer was spin-coated at 700 rpm and annealed at 110 °C for 30 min. The thickness of the active layer was measured with a Sloan Dektak II profilometer to range from 120 to 130 nm. The top and bottom electrodes of the devices were cleaned with a cotton stick soaked with chloroform to make contacts. Finally, 5 nm of MoO₃ and 100 nm of Ag were deposited with deposition rates of 0.2 Å/s and 1.0 Å/s, respectively, on all the devices using a physical vapor deposition system (PVD) under high vacuum at a pressure of 7×10^{-6} mbar. The active area of the devices was 6 mm². The inverted device structure and energy alignment is shown in Fig. 1a–b. The molecular structures of the active layer components are shown in Fig. 1c.¹

2.3. Characterization

X-ray diffraction (XRD) patterns were obtained using an Xpert Pro Philips powder X-ray diffractometer. UV–visible absorption spectra were taken using an Olid[®] HP8452 Diode Array Spectrometer. Fluorescence and photoluminescence (PL) spectra were measured with a USB2000-Ocean Optics spectrometer. The cyclic voltammograms were recorded using an EG&G Model 263 potentiostat connected with three electrodes: working electrode (glassy carbon-carbon), reference electrode (Ag/AgCl (in 3 M NaCl)) and counter or auxiliary electrode (Pt) in 0.1 M tetrabutylammonium hexafluorophosphate in dichloromethane at a scan rate of 50 mVs⁻¹. The morphology of ternary blend films and control device films were characterized using an Ambios multimode Atomic Force Microscope (AFM) in tapping mode with a 300 KHz resonant frequency cantilever. Current density-voltage (J-V) measurements were carried out using a Keithley 4200-SCS in the dark and under illumination. All the photovoltaic parameters of the inverted cells were measured under ambient conditions using a Xenon light with an intensity of 100 mW cm⁻² calibrated with an AM 1.5 solar simulator. Incident photon to electron conversion efficiency (IPCE) measurements were performed in the range between 300 and 800 nm

¹ Marvin was used for drawing, displaying and characterizing chemical structures, substructures and reactions, Marvin 15.1.12, 2015, ChemAxon (www.chemaxon.com).

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