



# Effect of blend composition on ternary blend organic solar cells using a low band gap polymer



Matthew Wright\*, Rui Lin, Murad J.Y. Tayebjee, Binesh Puthen Veettil, Yu Jiang, Xueting Liang, Ashraf Uddin, Gavin Conibeer

School of Photovoltaic and Renewable Energy Engineering, University of New South Wales, 2052 Sydney, Australia

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## ABSTRACT

This work investigates the influence of blend composition in ternary blend bulk heterojunction organic solar cells composed of poly(3-hexylthiophene-2,5-diyl) (P3HT), poly[2,1,3-benzothiadiazole-4,7-diyl [4,4-bis(2-ethylhexyl)-4H-cyclopenta [2,1-b:3,4-b']dithiophene-silole 2,6-diyl]] (Si-PCPDTBT) and [6,6]-phenyl C<sub>71</sub> butyric acid methyl ester (PC<sub>71</sub>BM). The use of the low band gap Si-PCPDTBT vastly improves the spectral response, when compared to a P3HT:PC<sub>71</sub>BM binary system. The optimum blend composition occurred at a total polymer concentration of 50 wt%. At low polymer concentrations, the P3HT phase was amorphous in nature. Increasing the polymer content led to the formation of crystalline polymer domains, as evidenced by XRD measurements. This significantly enhanced the charge carrier transport throughout the active layer. XPS depth profiles indicated that variations in the polymer content also influenced the mixing between the Si-PCPDTBT and the P3HT host matrix. This analysis showed that the 50 wt% was conducive to a larger interaction between the two polymers. A comprehensive analysis of the relative contributions of each molecule to the photoluminescence suggested that the polymer concentration not only affects the film microstructure, it also influences the photoluminescence quantum yield of the blend. This is caused by alterations to the recombination mechanisms occurring in the constituent materials, which, in turn, influences photocurrent generation. This result shows that the overall polymer content must be chosen carefully to strike a delicate balance between improved absorption and effective charge generation and collection.

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## 1. Introduction

Organic solar cells (OSCs) are envisioned to provide a low-cost alternative to conventional energy technologies, to help overcome rising global energy demands [1]. Extremely short energy payback times have been predicted for this technology [2], owing to the fast processing provided by solution printing and coating techniques. Multiple successful studies demonstrating OSCs in the field have focussed on the polymer, poly(3-hexylthiophene-2,5-diyl) (P3HT) [3,4]. P3HT is the most understood polymer in terms of device physics and physical stability [5].

Although the P3HT:PCBM system possesses many desirable traits, the wide optical band gap of P3HT (1.9 eV) places an inherent limit on the maximum achievable efficiency [6]. Due to the large band gap, the maximum theoretical photocurrent density for a binary blend OSC using P3HT is 19.7 mA/cm<sup>2</sup> [7], assuming that all

incident photons, with energies larger than the band gap, generate photocurrent. Due to the molecular nature of the polymer absorption and charge transport losses, the practically obtainable photocurrent is much lower than this [8,9]. The maximum efficiency achieved for a binary P3HT:PCBM OSC is 5–6% [10]. One potential solution to overcome this problem is to use a tandem architecture, whereby two solar cells, consisting of binary blends containing different polymers, can be connected, usually in series [11]. When connected in series, this cell architecture can exhibit high efficiencies, owing to large open circuit voltage ( $V_{oc}$ ), which can approach the sum of the  $V_{oc}$  values of the subcells [12]. However, this significantly complicates the fabrication process as it requires current matching of the sub-cells and a carefully designed 'recombination layer' to allow for lossless charge extraction [13]. Additionally, the number of layers in the device is significantly increased, which increases the complexity of device processing.

An alternative method to overcome this problem is to incorporate two separate polymer donor materials into a single junction device, forming a ternary blend solar cell [9,14,15]. Using two different polymers which have complementary absorption

\* Corresponding author.

E-mail address: [matthew.wright@unsw.edu.au](mailto:matthew.wright@unsw.edu.au) (M. Wright).

profiles allows for more efficient light harvesting in a single layer device, whilst only requiring relatively simple processing conditions, when compared to organic tandem cells. Koppe et al. demonstrated this concept by incorporating the low band gap polymer PCPDTBT ( $\sim 1.5$  eV) into a P3HT:PCBM host system [16]. The incorporation of a small amount of PCPDTBT led to sensitized absorption in the near infra-red region, at wavelengths where the P3HT:PCBM host system is not responsive. EQE measurements showed that this absorption in the PCPDTBT was efficiently converted to photocurrent, causing an improved short circuit current density ( $J_{sc}$ ) in the ternary blend. However, when large amounts (more than 20 wt% of the total polymer) of PCPDTBT were added to the blend, the performance was significantly reduced, due mainly to a rapid reduction in fill factor [16,17]. Li et al. determined the phase diagrams for P3HT:PCPDTBT:PCBM ternary blends to study the mixing behaviour of the sensitizer [18]. Whilst P3HT is a semi-crystalline polymer, PCPDTBT displays very little crystallinity. Ternary phase diagrams, constructed from differential scanning calorimetry (DSC), showed that the addition of PCPDTBT significantly reduced the crystallinity of the blend. Adding 20 wt% PCPDTBT caused an abrupt transition from semi-crystalline to amorphous. This alteration in the film microstructure correlated with a sharp drop in the device FF. This rapid deterioration in performance implies that using amorphous polymers as sensitizers in ternary blends may not be suitable for achieving high efficiencies. Ameri et al. investigated the influence of incorporating the silicon bridged analogue of PCPDTBT, Si-PCPDTBT, to a P3HT:PCBM host system [19]. By varying the P3HT:Si-PCPDTBT ratio, they showed that much higher loadings of this polymer can be introduced to the P3HT:PCBM host system whilst still maintaining a high fill factor. This allowed for very efficient sensitisation at long wavelengths. They speculate that the reason for the higher allowable Si-PCPDTBT concentrations is because Si-PCPDTBT, unlike PCPDTBT, does not disrupt the crystalline P3HT domains. This was further studied by Koppe et al. in a recent communication [20]. Using time-resolved pump-probe spectroscopy, they show that the transfer of positive charge from Si-PCPDTBT to the P3HT matrix is very efficient, even for high Si-PCPDTBT loadings. Coupled with the observation that Si-PCPDTBT does not significantly disrupt the P3HT crystallinity, this explains the effective sensitisation of the P3HT:PCBM system using Si-PCPDTBT. Most studies on ternary blends using P3HT as the host matrix keep the total polymer:fullerene ratio constant at 1:1. Due to the complex nature of the ternary blend consisting of two polymers, this may not be the blend composition which provides optimal light absorption, charge transfer and charge carrier transport throughout the active layer. Further understanding of the mechanisms determining the optimal blend composition for this promising ternary material system is required.

In this investigation, the total polymer to fullerene ratio was varied for P3HT:Si-PCPDTBT:PC<sub>71</sub>BM OSCs. The fullerene derivative PC<sub>71</sub>BM was used in this study, as it contributes more photocurrent density than PC<sub>61</sub>BM, due to increased light absorption [21,22]. For

this ternary system, the optimum device performance was achieved when a total polymer concentration of 50 wt% was used. At very low polymer contents, the polymer was not able to form crystalline domains, which led to very poor hole transport throughout the photoactive layer. Increasing the polymer loading led to increased crystallinity and improved charge transport. For very high polymer loadings, significant topographical changes in the film were observed, leading to poor metal contact formation. Additionally, XPS analysis indicated that mixing between Si-PCPDTBT and the P3HT phase is dependent on the blend composition and is maximised for the 50 wt% case.

## 2. Experimental

### 2.1. Materials and fabrication

Si-PCPDTBT and PC<sub>71</sub>BM were purchased from 1–Material and used as received. P3HT was purchased from Sigma–Aldrich. Patterned indium tin oxide (ITO) glass substrates (12 × 12 mm) were purchased from Lumtec. Devices were fabricated using an inverted architecture. The structure used was ITO/ZnO/P3HT:Si-PCPDTBT:PC<sub>71</sub>BM/MoO<sub>3</sub>/Ag. Before device fabrication, the ITO glass substrates were cleaned by ultrasonication in deionised water, acetone and isopropanol, each for 10 min. The first layer deposited was the ZnO electron transport layer. Full experimental details regarding the preparation of the sol–gel derived ZnO layer are described elsewhere [23]. The ternary system investigated in this report was P3HT:Si-PCPDTBT:PC<sub>71</sub>BM. The blend composition of the ternary system was systematically varied. Referring to previous reports [19], the P3HT/Si-PCPDTBT ratio was kept constant at 7/3. In this report, the total amount of polymer (P3HT+Si-PCPDTBT), by wt%, was varied from 20–80 wt%. The solution concentrations used in this report are summarised in Table 1. The casting solvent for all cases was chlorobenzene. All solutions were stirred overnight on a hotplate at 40 °C in a N<sub>2</sub>(g) purged glovebox. Next, the active layer was spin coated on top of the ZnO. For all cases, the active layer was annealed at 140 °C for 5 min, in a N<sub>2</sub> atmosphere. A 10 nm thick MoO<sub>3</sub> buffer layer was then thermally evaporated at a rate of 0.6 Å/s. Device fabrication was completed by the thermal evaporation of a 100 nm silver (Ag) electrode at a background pressure of 5 × 10<sup>-6</sup> mbar. The device area used was 0.13 cm<sup>2</sup>. For absorption and photoluminescence (PL) measurements, the active layer films were spin coated onto cleaned ITO glass substrates, without any buffer layers or metal electrode. Charge carrier mobilities were measured by fabricating hole- and electron-only devices [24,25]. Details regarding the structure and fabrication processes of hole- and electron-only devices are reported elsewhere [26].

### 2.2. Characterisation

All characterization was performed in air within two days of device fabrication. Absorbance was measured using a PerkinElmer

**Table 1**

Summary of the blend compositions investigated in this report. The total polymer content was varied by wt% from 20 to 80 wt%. Note that the P3HT:Si-PCPDTBT ratio remained constant at 7:3 for all ternary blends.

Total polymer content (wt%)	P3HT (mg)	Si-PCPDTBT (mg)	PC <sub>71</sub> BM (mg)	Blend composition	Solvent
Reference	10	0	10	50:0:50	0.5 ml chlorobenzene
20	2.8	1.2	16	14:6:80	
30	4.2	1.8	14	21:9:70	
40	5.6	2.4	12	28:12:60	
50	7	3	10	35:15:50	
60	8.4	3.6	8	42:18:40	
70	9.8	4.2	6	49:21:30	
80	11.2	4.8	4	56:24:20	

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