



Active layer thickness effect on the recombination process of PCDTBT:PC₇₁BM organic solar cells

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ABSTRACT

We investigated the effect of active layer thickness on recombination kinetics of poly [N-9'-hepta-decanyl-2,7-carbazole-alt-5,5-(4',7'-di-2-thienyl-2',1',3'-benzothiadiazole)] (PCDTBT) and [6,6]-phenyl C71-butyric acid methyl ester (PC₇₁BM) based solar cells. Analysis of the fitted Lambert W-function of illuminated current density–voltage (*J*–*V*) characteristics revealed increased recombination processes with increased active layer thicknesses. The ideality factor extracted from PCDTBT:PCBM solar cells continuously increased from 1.89 to 3.88 when photoactive layer thickness was increased from 70 to 150 nm. We found that such increase in ideality factor is closely related to the defect density which is increased with increased photoactive layer thickness beyond 110 nm. Therefore, the different density of defect states in PCDTBT:PCBM solar cells causes the different recombination paths where solar cells with a thicker active layer (≥ 110 nm) are considered to undergo coupled trap-assisted recombination processes while single-defect trap-assisted recombination is dominant for thinner (70–90 nm) PCDTBT:PCBM solar cells. As a result, we found that the optimal efficiencies of PCDTBT:PC₇₁BM solar cells were limited to the active layers between 70 and 90 nm. Particularly, when PCDTBT:PC₇₁BM solar cells were optimized with an active layer thickness of 70 nm, energy conversion efficiency reached 6.5% while an increase in thickness led to the reduction of efficiency to 4.7% at 133 nm but then an increase to 5.02% at 150 nm.

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1. Introduction

Organic solar cell (OSC) technology has received significant attention over the past decade due to the simple, flexible nature of polymer photovoltaics and the potential to develop a clean, cost-efficient renewable energy source. The key development of organic solar cells has been made with the pioneering concept of “bulk heterojunction (BHJ)”

photoactive layers [1,2]. Particularly, BHJ organic solar cells blend from randomly-networked polymer:fullerene phases with offset energy levels where extremely low charge-carrier mobility, slow hopping transport through intra-molecules, and limited diffusion lengths lead to high charge-carrier recombination rates [3–5]. Therefore, one of the greatest obstacles in organic photovoltaics that researchers face is ensuring perfect collection of charge carriers by the electrodes before undergoing recombination. One of the approaches to optimize the efficiency of polymer-based, planar organic solar cells is through nanoscale control of constituent layer materials composing the device [3–5]. Particularly, there are considerable reports in literature describing charge transport mechanism and its

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impact on the efficiency of bulk-heterojunction organic solar cells. Extensively studied organic solar cells composed of regioregular poly(3-hexylthiophene) (P3HT) and phenyl-C61(or C71)-butyric acid methyl ester (PCBM) organic blends [6,7] have been reported. However, even though P3HT:PCBM is well studied, the energy conversion efficiency is limited to less than 5% due to a lower open circuit voltage (V_{OC}) and short circuit current (J_{SC}) [6]. Recently, as an alternative, poly[N-9'-hepta-decanyl-2,7-carbazole-alt-5,5-(4',7'-di-2-thienyl-2',1',3'-benzothiadiazole)] (PCDTBT) and [6,6]-phenyl C71-butyric acid methyl ester (PC₇₁BM) based organic solar cells have received intensive study due to higher energy conversion efficiency of more than 6% [8,9]. This is due to a better alignment of molecular orbital energy levels between the PCDTBT polymer and the PCBM fullerene, resulting in a higher open-circuit voltage (V_{OC}) of more than 0.8 V [8,9]. Furthermore, the lower optical band-gap of PCDTBT (1.8 eV) compared to P3HT (1.9 eV) is more beneficial to increase photon absorption compared to P3HT at longer wavelengths [8]. However, it is reported that PCDTBT:PCBM based organic solar cells have decreased efficiencies with increased thicknesses [10,11]. Particularly, the highest efficiency of PCDTBT:PCBM was obtained with photoactive layer thickness of 70–90 nm [10]. One of the possible assumptions regarding such decrease in energy conversion efficiencies is increased series resistance and recombination processes involved in the photoactive layer. However, until now, energy conversion processes, particularly charge transport mechanism, of PCDTBT:PCBM solar cells with different active layer thickness have not been systematically studied. In this paper, we fabricated PCDTBT:PC₇₁BM organic solar cells with different active layer thicknesses from 70 to 150 nm. These solar cells showed corresponding energy conversion efficiencies ranging from 6.5% to 4.7% which is well developed for the studies of recombination processes with different active layer thickness. To elucidate the device performance of these solar cells, optical simulation using transfer matrix method (TMM) [12–14] was used to predict the generation rate of electron-hole pairs in the active layer. Furthermore, the illuminated J - V curves were fitted with the well-known Lambert W-function [15] to extract the essential diode parameters to provide an insight into the recombination processes governing PCDTBT:PC₇₁BM organic solar cells. Based on these results, charge transport models have been developed to explain trap-assisted recombination processes of PCDTBT:PCBM solar cells fabricated with different active layer thicknesses.

2. Experimental details

PCDTBT polymer ($M_w \sim 59$ k, PDI ~ 1.8) was synthesized by following the synthetic route of Leclerc's group [16]. PC₇₁BM acceptor was purchased from Nano-C and used as received. Bulk heterojunction (BHJ) solar cells were fabricated as follows. First, ITO-sputtered glass substrates were cleaned, ultrasonicated in deionized (DI) water, acetone, and IPA, and dried overnight at 80 °C. The dried ITO substrates were transferred into an UV-ozone (UVO) clea-

ner to create a hydrophilic ITO surface. On ITO a hole transport layer (HTL) of PEDOT:PSS (AI 4083 as purchased) was spin-cast at 5000 rpm and dried at 150 °C for 10 min in air. The HTL-coated substrates were transferred into a N₂-glove box for coating photoactive BHJ layers. The photoactive layers were spin-cast at 500–3000 rpm by using a blend solution of PCDTBT (7 mg/ml) and PC₇₁BM (28 mg/ml) dissolved in 1,2-dichlorobenzene (o-DCB). The BHJ films were dried at 60 °C (40 min) to remove the solvent. An electron transport layer (ETL) of titanium sub-oxide (TiO_x) was spin-cast at 5000 rpm on the photoactive layer, in which a 300 times diluted TiO_x precursor solution with IPA was used for fabricating a thin 7 nm thickness TiO_x layer [17]. To deposit a top cathode metal the samples were transferred into a vacuum chamber, and aluminum (Al) was thermally evaporated at $<10^{-7}$ torr, producing a device structure consisting of glass/ITO (100 nm)/PEDOT:PSS (25 nm)/PCDTBT:PCBM (70–150 nm)/TiO_x (7 nm)/Al (80 nm). The device active area was defined by cathode area (14.5 mm² as measured). Current density–voltage (J - V) characteristics were measured within a N₂-glove box by using a Keithley 238 source measurement unit under air mass 1.5G solar illumination (100 mW/cm²). Incident photon-to-current efficiency (IPCE) spectra were measured by using a PV Measurement IPCE system (PV measurements, Inc.).

3. Results and discussion

PCDTBT:PC₇₁BM organic solar cells were carefully designed to scrutinize the effect of active layer thickness on the performance of devices. Particularly, the photoactive layer thicknesses of PCDTBT:PC₇₁BM organic blends were varied from 70 nm to 150 nm. Fig. 1a shows resultant current density–voltage (J - V) characteristics of PCDTBT:PC₇₁BM solar cells measured under AM 1.5G illumination (100 mW/cm²). As can be seen in Fig. 1a, the variation of the photoactive layer thickness had a marked impact on the J_{SC} and, to a lesser extent on the V_{OC} . Interestingly, the J_{SC} declined from 70 nm to 133 nm but then once again increased at 150 nm. Fig. 1b shows incident photon to charge carrier efficiency (IPCE) which dominated at wavelengths between 350 nm and 550 nm for all devices. The maximum IPCE was achieved over 71% for PCDTBT:PC₇₁BM devices with active layer thicknesses of 70 nm and 150 nm, indicative of efficient photon-to-electron conversion process. On the contrary, organic solar cells with active layer thicknesses of 90, 110, and 133 nm had maximum IPCE of less than 70%. Therefore, IPCE showed a similar trend that was observed in J_{SC} as shown in Fig. 1a.

To qualitatively analyze such tendency observed in J_{SC} and IPCE, we conducted optical simulation using a transfer matrix method (TMM) [12–14] which allows for analytically describing light propagation by taking into account the cumulative effects of reflection and transmission at all interfaces and absorption in each layer of the multilayered system. Particularly, the distribution and the rate of exciton generation were calculated by numerically solving a number of matrix equations in the multilayers. For each of the layered materials the complex indices of refraction

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