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Composition-dependent phase separation of P3HT:PCBM composites for high performance organic solar cells

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ABSTRACT

Phase separation of the poly(3-hexylthiophene) (P3HT) and [6,6]-phenyl-C_{61}-butyric acid methyl ester (PCBM) active layer was investigated by varying its relative ratio in the bulk heterojunction (BHJ) organic solar cells (OSCs). The UV/visible absorption spectra of P3HT and PCBM were proportional to their compositions in the active layer. However, the crystallinity of P3HT, characterized by X-ray diffraction (XRD), improved with increasing composition up to 50 wt.%, but then degraded upon any further increase in its composition above 50 wt.%. This unique phenomenon corresponded to the microstructure of the layer, which was analyzed using transmission electron microscopy (TEM). The fraction of the highly ordered fiber structure of P3HT increased as the P3HT composition increased up to 50 wt.% and decreased as the composition increased further, which was in accordance with the XRD results. The microstructure of the P3HT:PCBM active layer, determined by the composition-dependent phase separation, supported the optimized performance of the OSCs with the active layer composition of 50% P3HT and 50% PCBM.

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1. Introduction

Bulk heterojunction (BHJ) based organic solar cells (OSCs) with composites made of a conjugated polymer and fullerene derivative active layer have considerable potential as renewable energy resources. From a material point of view, blends of poly(3-hexylthiophene) (P3HT) and [6,6]-phenyl-C_{61}-butyric acid methyl ester (PCBM) are most widely used as an active layer and have the highest recorded power conversion efficiency (PCE) up to 5% thanks to the thermal and illuminative stability [1] and high electron mobility [2]. It can also be fabricated on a large area substrate using a simple solution process because of its high solubility in organic solvents [3,4]. The microstructure of the donor and acceptor BHJ should have a large interface area with nanometer-scale dimensions in order to guarantee the efficient charge separation for highly efficient OSCs because the typical diffusion length range of exciton is 4–20 nm in polymers [5].

Since the performance of the OSCs is determined by the properties of the P3HT:PCBM active layer and its microstructure, there have been intensive studies on the optical absorption, chemical properties, phase separation and consequent microstructure of the P3HT:PCBM active layer [6–14]. The UV/visible absorption peak of P3HT red-shifts to a lower energy state and is broader and more vibronic as its structure becomes more ordered with a longer chain [6–9]. Additionally, reports have shown that the performance of the OSCs improved through the formation of a highly ordered nano-scale structure with a large interface area between P3HT and PCBM [10,11]. The effect of the processing conditions such as the kind of solvent, annealing time, and temperature has been investigated for phase separation of the active layer in order to form the highly

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ordered structure of the active layer [10–12]. The composition of the P3HT:PCBM active layer, i.e. the relative ratio of each polymer, also influences the microstructure of the active layer and consequently the performance of the OSCs [9,12]. Several research groups have reported that the optimized morphologies and photovoltaic (PV) performances occurred at a 1:1 ratio of the P3HT:PCBM (50 wt.% of P3HT) composites [11,13]. Recently, reports have shown that the use of a low composition (1:0.8–0.7 ratio of P3HT:PCBM) of PCBM resulted in a PCBM cluster free structure and thermally stable devices [1,14]. Although there have been several reports about the effect of the composite ratio on the performance of the OSCs mostly with the optical and electrical properties [9,12–14], further studies are required in order to understand the relationship between the properties and the microstructure of the active layer associated with phase separation depending on the ratio of P3HT:PCBM and the OSCs performance.

In this study, the effect of the P3HT:PCBM composition in the active layer was studied by varying the composite ratio of P3HT from 20 to 80 wt.%. The optical properties and microstructure of the active layer and the correlation with the device performance were explored.

2. Experimental

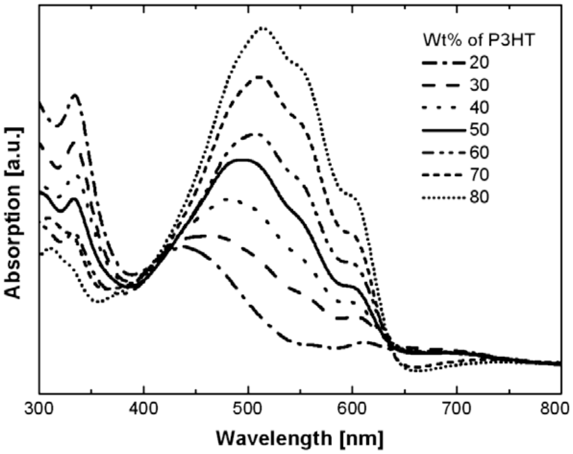
Regioregular P3HT (Rieke Metals, Inc.) and PCBM (Nano-C) were used for the active layer as purchased without any further purification. P3HT and PCBM were separately dissolved in chlorobenzene (30 mg/ml for each solution) and stirred at 60 °C for 1 h. These solutions were mixed with different weight ratios of: 20, 30, 40, 50, 60, 70, and 80 wt.% of P3HT. The substrate for the OSCs, the indium tin oxide (ITO; 10 Ω/square) coated glass substrate, was cleaned in an ultrasonic bath with deionized water, acetone and isopropyl alcohol for 15 min each, and then cleaned with UV–ozone for 10 min. The poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS; Baytron P VP AI 4083, H.C. Stark), used as an anodic buffer layer, was spin coated at 6000 rpm for 10 s and baked at 130 °C for 10 min. The P3HT:PCBM photoactive layer was spin coated on the PEDOT:PSS at 4000 rpm for 5 s. The thicknesses of the buffer and active layers were 30 nm and 70 nm, respectively, as measured using a surface profiler (alpha-step 500). The Al (100 nm) cathode was thermally evaporated through a shadow mask defining active area of 0.1 cm^2. Thermal annealing was conducted at 150 °C in an oven for 10 min under ambient N_2. The entire fabrication process was conducted in an air environment except for thermal annealing.

An UV/visible spectrophotometer (Shimadzu UV-1601) was used to characterize the absorption properties of the P3HT:PCBM layer. The crystallization of the active layer was analyzed using X-ray diffraction (XRD; Mac Science, Cu Kα wavelength of 1.54056 Å). The phase separation and resulting microstructure were observed using optical microscopy and field-emission transmission electron microscopy (FE-TEM; JEM-2100F, JEOL). For the TEM analysis, the specimens were prepared by floating the active layers onto the deionized water and then picking them up with a Cu grid. The current density versus voltage (*J*–*V*) characteristics were measured with a Keithley 236 source measurement unit and a solar simulator (Yamashita denso) under an AM 1.5 G (100 mW/cm^2) irradiation intensity. The total incident light intensity was calibrated with a pyranometer (Eko MS-802) and a standard reference silicon solar cell.

3. Results and discussion

Fig. 1 shows the UV/visible absorption spectra of the P3HT:PCBM active layer with different concentration ratios. A previous reports showed that the maximum absorption wavelengths (λ_{max}) of PCBM molecules are almost constant regardless of the amount of the PCBM composite or ordering of the active layer [9,15,16]. In this study, the intensity of the PCBM absorption spectra (~330 nm) increased without any shift in the wavelength as the composition of PCBM increased. On the other hand, as the composition of P3HT increased, the absorption spectra of P3HT between 450 and 650 nm gradually increased, was broader as well as more vibronic, and red-shifted. The λ_{max} of the P3HT peak were at 437, 461, 482, 495, 507, 512 and 515 nm for 20, 30, 40, 50, 60, 70 and 80% of P3HT, respectively. The shift of absorption peaks to higher energy indicates an increasing density of conformational defects, equivalent to non-plarnarity, and causes loss of conjugation [17]. Moreover, the absorption shoulder at ~610 nm which is assigned to interchain interactions became bigger and more pronounced. Since the thickness of the active layer was kept the same (~70 nm) for the all of the samples, the increased, broadened, red-shifted and pronounced absorption peaks of P3HT were attributed not only to the increased composite ratio but also to the closer chain packing of P3HT which resulted from strong interchain interactions [9,12,17,18].

The XRD analysis in Fig. 2 depicted the crystallization of P3HT, which was represented by the peak intensity and the



Wt% of P3HT [%]	20	30	40	50	60	70	80
P3HT λ_{max} (nm)	437	461	482	495	507	512	515

Fig. 1. UV/visible absorption spectra of the active layer with various composite ratios of P3HT:PCBM after thermal annealing at 150 °C for 10 min in a N_2 flow oven.

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