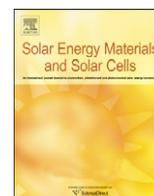




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Effects of a perfluorinated compound as an additive on the power conversion efficiencies of polymer solar cells

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ABSTRACT

A perfluorinated compound, 4-amino-2-(trifluoromethyl)benzotrile (**ATMB**), was applied as an additive to polymer solar cells (PSCs) with P3HT [poly(3-hexylthiophene)]:PCBM [[6,6]-phenyl-C₆₁-butyric acid methyl ester] blend films. The addition of 6 wt% **ATMB** to a P3HT:PCBM layer led to an increased power conversion efficiency of 5.03% due to the enhanced short circuit current and fill factor when compared with that of the reference cell without an additive. On the other hand, the devices with 4-aminobenzotrile as an additive, not containing fluorine atoms in the molecule, displayed lower PCEs than that of the reference cell. The UV–visible absorption spectra, X-ray measurements and carrier mobility studies revealed that **ATMB** facilitated ordering of the P3HT chains, resulting in higher absorbance, larger crystal size of P3HT and enhanced hole mobility. XPS depth profiling measurements also showed that the additive molecules were predominantly positioned in the range of 25 nm under the surface of the P3HT:PCBM film, leading to improved fill factor.

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1. Introduction

Polymer solar cells (PSCs) are a promising and new renewable resource for the generation of electrical energy primarily due to their low manufacturing costs [1,2]. For example, roll-to-roll processed PSCs have been fabricated onto flexible substrates with larger areas using flexographic printing, slot die coating and rotary screen printing [3–7], and their modules have been applied to charge electronic products such as white light emitting diode and battery [4,5]. Recently, the power conversion efficiency (PCE) of PSC has been improved by adopting a bulk hetero-junction (BHJ) structure composed of poly(3-hexylthiophene) (P3HT, electron donor) and [6,6]-phenyl-C₆₁-butyric acid methyl ester (PCBM, electron acceptor) [2,8,9]. However, this efficiency is not sufficient for the realistic specifications of the commercial utilization of PSCs. The main limiting factors for the PCE of PSCs are poor exciton/charge transport and narrow absorption in the visible range of the solar spectrum in the active layer. The donor and acceptor materials should form nano-scaled interpenetrating networks within the whole active layer to ensure efficient exciton dissociation and charge transport [10]. Therefore, significant studies to improve light harvesting, photocurrent generation and exciton/charge transport in the active layer have had a

morphological basis. Several interesting attempts, such as thermal annealing at a specific temperature [11,12], controlling the solvent evaporation rate [9], incorporating processing additives [13] or non-volatile additives into active layer [14] and controlling the charge injection at the organic–inorganic interface [15,16], have been used to obtain nano-scaled bicontinuous morphology. However, among these methods, the additive technique together with post-thermal annealing is regarded as the best method to create nano-sized phase-separation in the active layer and molecular structural ordering within the nano-domains due to its easy applicability [17]. Additionally, organic semiconductors, for example, 3-hexyl thiophene [18], Cu⁺ chelated polyalkoxythiophene [19], phthalocyanine derivatives [20] and 9,10-diphenylanthracene [21], have also been applied to PSCs as processing additives to control morphologies of the active layers. It was also proven that liquid crystalline (LC) materials are also effective processing additives in P3HT:PCBM-based PSCs [22,23].

Meanwhile, it was reported that fluorine compounds induce an increase in PCE of BHJ solar cells. When polyvinylidene fluoride (PVDF) was applied as an interlayer between P3HT:PCBM blend film and poly-3,4-ethyleneoxythiophene:poly-4-sytronsulfonate (PEDOT:PSS) buffer layer, extensive phase separation and high crystallization of the P3HT:PCBM blend film attributed to the hydrophobicity of the PVDF film were achieved [24]. The incorporation of a newly synthesized fullerene derivative with a perfluoroalkyl group into the P3HT:PCBM blend film largely enhanced the fill factor due to the spontaneous formation of thin

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buffer layer [25]. Chen et al. [26] recently reported that fluorine-containing low band gap polymer (PBDTTT-CF) showed a PCE of over 7%, much superior to those of other thienothiophene derivatives without fluorine atoms.

In this study, we adopted a perfluorinated compound [4-amino-2-(trifluoromethyl)benzonitrile—*ATMB*] as an additive, which is widely used for the synthesis of LC materials and mass-produced by many manufacturers in the world, for application to P3HT:PCBM-based PSCs to improve device efficiency. We expected that the very simple addition of the perfluorinated compound into P3HT:PCBM blend film possibly works as a medium to enhance photovoltaic performance as in previous literatures. PSCs containing additives were fabricated and their photovoltaic properties are discussed.

2. Experimental

2.1. Materials

A glass substrate with a patterned indium–tin oxide (ITO, $10 \Omega/\square$) layer for PSCs was purchased from SUNIC Ltd. (Korea). 98% Regioregular P3HT and PCBM were purchased from Rieken Metals and Nano-C, respectively. The weight-averaged molecular weight of P3HT was 6.4×10^4 g/mol. The perfluorinated compound (*ATMB*) and 4-aminobenzonitrile (ABN) as a counter material were purchased from Tokyo Chemical Industry Co., Ltd. and used without further purification. The chemical structures of *ATMB* and ABN used as additives in this study are shown in Fig. 1. PEDOT:PSS (Baytron P VP Al 4083) was used as received from H.C. Stark GmbH.

2.2. Device fabrication

PSCs were fabricated on ITO glasses as follows. ITO glasses were cleaned by ultrasonic treatment in acetone, detergent, de-ionized water and isopropyl alcohol sequentially and then dried in a vacuum oven. Next, O_2 plasma treatment on ITO glasses was conducted for 3 min and immediately afterwards, PEDOT:PSS was spin-coated on the ITO glass. The PEDOT:PSS films were then baked on a hot plate for 15 min at 140°C to give a thickness of 30 nm and moved to a N_2 glove box for the remainder of the fabrication process. P3HT and PCBM with weight ratio of 1:0.9 were first dissolved in chlorobenzene. Various amounts of additives, *ATMB* or *ABN*, (ranging from 1 to 10 wt% based on P3HT:PCBM) were then added to the P3HT:PCBM solutions, followed by stirring for 24 h at 50°C . The solution consisting of P3HT:PCBM blended with or without additives was spin-cast on the top of the PEDOT:PSS layer, and the plate was then dried on a hot plate in a covered Petri dish for 40 min at 50°C to produce an active layer with a thickness of 100 nm. Finally, a cathode consisting of LiF (1 nm) and a subsequent Al (150 nm) layer was deposited by thermal evaporation under a vacuum of 10^{-7} Torr. The devices with the configuration of ITO/PEDOT:PSS (30 nm)/

P3HT:PCBM:with or without additive (100 nm)/LiF (1 nm)/Al (150 nm) were encapsulated with a glass cap to protect them from air. The active area of all devices was defined as 9 mm^2 by a shadow mask. Hole-only devices [27] were fabricated with a diode configuration of ITO/PEDOT:PSS (30 nm)/P3HT:PCBM:with or without additive (100 nm)/Au (100 nm), where Au with a high work function was used as an electrode to maintain the injection of electrons. By replacing PEDOT:PSS with a Cs_2CO_3 layer [28] spin-coated from its solution in 2-ethoxyethanol, electron-only devices were fabricated with a diode configuration of ITO/ Cs_2CO_3 /P3HT:PCBM:with or without additive (100 nm)/LiF (1 nm)/Al (150 nm) for the measurement of electron mobility.

2.3. Characterization

UV–visible absorption spectra of the fabricated blend films with and without additive were obtained using a Perkin Elmer Lambda 750 UV/VIS spectrometer. Reflectance spectra were recorded with a Konica Minolta Spectrometer (CM-3600D). The X-ray diffraction (XRD) profiles were measured by a BRUKER D8 ADVANCE with Cu $K\alpha$ as the incident beam. Data were obtained from 3° to 30° (2θ) at a scan rate of 0.2 s/step. X-ray photoelectron spectroscopy (XPS) was applied to detect fluorine atoms in the active layer using a Kratos Axis Ultra XPS system with a base pressure of 1×10^{-10} mbar using a monochromated Al $K\alpha$ X-ray source. XPS survey scans were taken at a pass energy of 160 eV. Photocurrent–voltage measurements were performed with a Keithley model 2400 SourceMeter and a Newport 91192 solar simulator system (equipped with 1-kW Xenon arc lamp from Oriel). The light intensity was adjusted to simulated AM 1.5 radiation at 100 mW cm^{-2} with a Radiant Power Energy Meter (Model 70260, Oriel). IPCE (incident photon to current conversion efficiency) results were acquired from an IPCE G1218a (PV measurement). This system applied monochromatic light from a 75-W Xenon arc lamp (Ushio UXL-75XE), which was filtered by a dual-grafting monochromator and an individual filter onto the test devices. An all-ellipsoidal reflector collected light from the lamp and focused it on the monochromatic entrance slit via a mechanical chopper to create a small modulated signal. While modulated, monochromatic light was applied to the test devices, continuous bias light (ca. 1 sun) was also applied. All measurements were carried out under ambient conditions at room temperature.

3. Results and discussion

3.1. Optimization of additive contents

The PSCs with the configuration of ITO (150 nm)/PEDOT:PSS (30 nm)/P3HT:PCBM:with or without additive (100 nm)/LiF (1 nm)/Al (150 nm) were fabricated and characterized before and after post-thermal annealing for 25 min at 145°C . To achieve the highest efficiency, the amount of additives, *ATMB* or *ABN* was varied from 0 to 10 wt%. The resulting photovoltaic properties of the devices characterized under AM 1.5 conditions are presented in Fig. 2. The performance of the as-prepared PSCs displayed relatively poor photovoltaic properties regardless of the amount (ranging from 0 to 10 wt%) of *ATMB* or *ABN*. After post-thermal annealing, the performance of devices with *ABN* additive was much inferior to that of the control device (An-PSCO) without *ABN* as shown in Fig. 2d. From the sharp decrement in J_{sc} (short circuit current) compared to that of the control device, *ABN* additives do not seem to affect the formation of P3HT and PCBM nano-domains during post-thermal annealing. On the other hand, the performance of annealed devices with incorporation of 3–6 wt% *ATMB* considerably improved, compared with the device

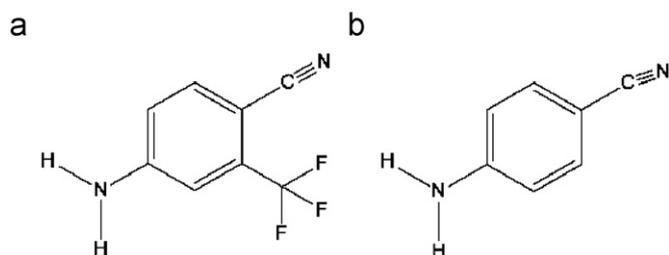


Fig. 1. Chemical structures of (a) *ATMB* and (b) *ABN*.

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