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Influence of alkanediol series as processing additives in photo-active layer on the power conversion efficiency of polymer solar cells

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ABSTRACT

The alkanediol series such as 1,2-ethanediol, 1,3-propanediol, 1,4-butanediol, 1,6-hexanediol, 1,8-octanediol and 1,10-decanediol was introduced to poly(3-hexylthiophene, P3HT):[6,6]-phenyl-C₆₁-butyric acid methyl ester (PCBM) based bulk-heterojunction (BHJ) polymer solar cells as processing additives. When 1,3-propanediol was introduced to photo-active layer as a processing additive, power conversion efficiency (PCE) was dramatically increased from 3.5% (for the reference device) to 5.3%. To investigate the causes of improvement of the PCE of PSCs after addition of alkanediol series, photon absorbance properties were observed using UV–vis spectroscopy and photoluminescence (PL). Then, the degree of crystallinity of P3HT was examined through X-ray analysis. In addition, changes in the nano-structure and surface morphology of P3HT:PCBM blend film were observed through AFM and TEM. The series resistance (R_s) of the fabricated PSCs was 5.19 Ω cm², and short circuit current density (J_{sc}), open circuit voltage (V_{oc}) and fill factor (FF) were 16.2 mA/cm², 0.63 V and 51.9% respectively.

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1. Introduction

Thanks to their good stability for thin film formation, good mechanical properties and excellent electro-optical properties, π -conjugated polymers have attracted great attention as next-generation device materials for organic electric devices [1–5]. In polymer solar cells (PSCs), in particular, solution processes such as screen printing [6], ink-jet printing [6], slot die coating [7], roll to roll stamping [8–11] and brush painting method [12,13] are applicable. Therefore, light-weight and flexible large-area photo-voltaic devices can be implemented at low costs. Typically, bulk heterojunction (BHJ) PSCs, which are promising device configuration for high power conversion efficiency (PCE), involve the use of a phase-separated blend of an electron-donating material (p-type conjugated polymers) and an electron-accepting material (n-type fullerene derivatives) as the active layer [14,15].

Tremendous efforts have been made to optimize the photo-active layer formation [16] and the device configuration [17,18], as the maximum PCE of the BHJ PSCs are dramatically increased up to about 8–9% [19,20]. To improve the PCE of BHJ PSCs, it is necessary to properly form continuous pathways to transfer excited electron and hole to the external circuit without leading to their recombination with each other. The typical BHJ PSCs constitute the system which has poly(3-hexylthiophene, P3HT) as

an electron donor and [6,6]-phenyl-C₆₁-butyric acid methyl ester (PCBM) as an electron acceptor. In this system, PCBM is dispersed among P3HT chains and prevents P3HT from being crystallized. Therefore, initial BHJ PSCs show 2.8% of PCE [21]. In order to improve the PCE of P3HT/PCBM BHJ PSCs, therefore, P3HT and PCBM should exist in an interpenetrating network form, and they should be phase-separated very well. In several groups, efficiency has been improved by forming a well-defined bicontinuous interpenetrating network between P3HT and PCBM by controlling thermal annealing time and temperature of BHJ films and using various solvents for dissolving P3HT and PCBM [22–30]. Due to these efforts, P3HT based PSCs improved PCE up to ca. 5%.

As an alternative, Zhang et al. introduced ‘solvent mixture’ method which blended two different solvents in terms of boiling point [31]. Even though P3HT chain was reorganized by introducing the solvent mixture to which a small amount of chlorobenzene (b.p. 132 °C) was added to the host solvent ‘chloroform (b.p. 61 °C)’, low PCE (2.3%) was observed.

The studies on BHJ PSCs with low-band gap polymers as an electron donor material whose performance was enhanced by adding a few volume percent of processing additives (e.g. 1,8-diiodooctane, 1,8-octanedithiol, etc.) to the host solvent in a similar method have also been reported [32,33]. These processing additives can improve the order in low-band gap polymer blends with PCBM. The improved order was reflected in a higher hole mobility, higher FFs, and a reduced series resistance. However, most known processing additives have been applied to the cases in which low-band gap polymer exist.

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In P3HT/PCBM system, alkanethiols (e.g. n-hexylthiol, n-octylthiol or n-dodecylthiol.) were added to P3HT/PCBM blend, and these allowed the formation of thin films with slightly enhanced hole mobility and charge-carrier lifetimes, because of enlarged P3HT domains with higher crystallinity. Nevertheless, some thermal annealing was still necessary to give the highest possible performance [34].

In this study, therefore, PCE of PSCs was dramatically increased through improving the order in P3HT blends with PCBM and photon absorption property of BHJ film by introducing the novel processing additives 'alkanediol series' to the P3HT/PCBM system. Alkanediol series, which was used as a processing additive, has a higher b.p. than the host solvent as 1,2-dichlorobenzene (ODCB, 180 °C). In addition, we found that the alkanediol selectively dissolves PCBM which was well dispersed in alkanediol, but P3HT was not soluble in alkanediol. For this reason, adding alkanediol series to ODCB without any thermal treatment could improve the ordering of the polymer domain. This study has figured out the causes of improvement of the properties of BHJ PSCs by observing the optical, electrical and structural properties of BHJ film after addition of processing additives. Using UV/ PL spectroscopy, the absorption of photon and the quenching properties of excited electron were observed. In addition, the structural properties of P3HT crystalline were analyzed through XRD pattern analysis. Furthermore, phase segregation of P3HT/PCBM film and increase of crystallinity in P3HT were observed through AFM and TEM analyses.

2. Materials and measurements

2.1. Materials

Indium tin oxide (ITO) glass used as the transparent electrode was produced by Samsung Corning (ITO:170 nm, 10 Ω/sq). Poly(3,4-ethylenedioxythiophene)/poly(styrenesulfonate) (PEDOT:PSS, Al 4083) was purchased from Clevios, and P3HT, which was used as a donor material in the photoactive layer, was purchased from Rieke metal. PCBM, the acceptor material, was purchased from Nano C. 1,2-dichlorobenzene (ODCB), used as a host solvent, and 1,2-ethanediol (2-DO), 1,3-propanediol (3-DO), 1,4-butanediol (4-DO), 1,6-hexanediol (6-DO), 1,8-octanediol (8-DO) and 1,10-decanediol (10-DO), used as processing additives, were purchased from Aldrich. The chemical structures of P3HT and PCBM are shown in Fig. 1(a) and (b) respectively. In addition, the chemical structures of alkanediol series are shown in Fig. 1(c).

2.2. Measurements

All of the thin films were fabricated using a GMC2 spin coater (Gensys), and their thicknesses were measured using an alpha step 500 surface profiler (KLA-Tencor). The photon absorption property and the quenching property of the excited electrons in the BHJ film were measured with UV-vis spectroscopy (HP Agilent 8453) and PL spectroscopy (Perkin Elmer LS 55 luminescence spectrometer), respectively. The XRD patterns were observed using a Rigaku D/MAX 2200 diffractometer with Cu Kα radiation to confirm the crystalline size of P3HT. The morphology of the BHJ films was observed through AFM (PSIA XE-100) and FE-TEM (FEI Tecnai G2 F30), respectively. The surface energy was measured with a contact-angle meter (KRUSS K6). The current density–voltage (*J*–*V*) characteristics of the PSCs were measured using a Keithley 2400 source measure unit. The devices were evaluated at 298 K by using a Class A Oriel solar simulator (Oriel 96000 150 W solar simulator) with a xenon lamp that simulates

AM 1.5G irradiation (100 mw/cm²) from 400 to 1100 nm. The instrument was calibrated with a monocrystalline Si diode fitted with a KG5 filter to bring the spectral mismatch to unity. The calibration standard was calibrated by the National Renewable Energy Laboratory (NREL). IPCE (Mc science) was measured against the best-performing device.

3. Experimental part

3.1. Cleaning of patterned ITO glass

To clean the patterned indium tin oxide (ITO) glass, detergent (Alconox[®] in deionized water, 10%), acetone, isopropyl alcohol and deionized water were each sonicated in order for 20 min. Moisture was removed by blowing thoroughly with N₂ gas. To ensure complete removal of all of the remaining water, the patterned ITO glass was baked on a hot plate for 10 min at 100 °C. For hydrophilic treatment of the patterned ITO glass, it was cleaned for 10 min in a UVO cleaner.

3.2. Fabrication of PSCs

PEDOT:PSS was spin coated from aqueous solution to form a film of thickness of ca. 40 nm on the patterned ITO glass. The substrate was dried for 20 min at 120 °C in air and then transferred into a glovebox for spin coating the active layer. P3HT and PCBM were dissolved in ODCB with 1:0.6 (P3HT:PCBM=10 mg/ml:6 mg/ml) weight ratio. 2-DO, 3-DO, 4-DO and 6-DO were added to the solutions with various concentrations and mixed by a centrifugal mixer (Thinky ARE-310) at 2000 rpm for 10 min in order to make a well dispersed solution. 8-DO and 10-DO, which are white solid, were insoluble in ODCB; the mixture solution was subjected to thermal treatment by hot plate at 100 °C for 1 h. Then, mixture solution, in which processing additives as 8-DO and 10-DO were added, was mixed by a centrifugal mixer at 2000 rpm for 10 min. The blend solution was then spin coated on top of the PEDOT:PSS layer. The thickness

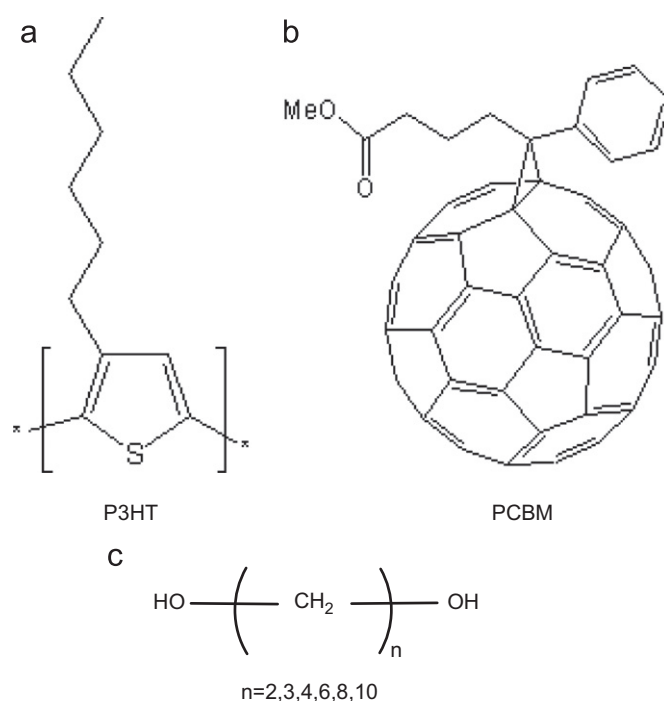


Fig. 1. The chemical structures of (a) P3HT, (b) PCBM and (c) alkanediol series as processing additives.

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