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A thienylenevinylene-phthalimide copolymer based polymer solar cell with high open circuit voltage: Effect of additive concentration on the open circuit voltage



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ABSTRACT

Polymer solar cells (PSCs) based on alkyl-substituted thienylenevinylene and phthalimide-based D-A copolymer (PTVPhI-Eh) as a donor and [6,6]-phenyl-C71-butyric acid methyl ester (PC₇₁BM) as an acceptor were investigated with the intent of gaining high performance photovoltaic properties. PTVPhI-Eh had a deep highest-occupied molecular orbital (HOMO) level, ~0.3 eV lower than rr-P3HT. As a result, PTVPhI-Eh:PC₇₁BM devices showed a high open circuit voltage (V_{oc}) of 0.87 V, which is over 0.2 V higher compared to that of P3HT:PCBM-based PSCs. The maximum power conversion efficiency (PCE) of PSCs was 4.14% with V_{oc} of 0.87 V, short circuit current (J_{sc}) of 7.32 mA/cm², and fill factor (FF) of 65%, under optimized conditions of a 1:1 wt/wt ratio and treatment with a 2 vol% 1-chloronaphthalene solvent additive. The V_{oc} and consequent photovoltaic performance of this PSC was significantly affected by the concentration of the solvent additive. To determine the relationship between V_{oc} and concentration of solvent additive, PTVPhI-Eh:PC₇₁BM devices were investigated with various analyses such as optical spectroscopy, transmission electron microscopy (TEM), and X-ray diffraction (XRD).

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1. Introduction

Polymer solar cells (PSCs), the so-called bulk heterojunction (BHJ) solar cells, have been of interest as promising technology for renewable energy because of the unique advantages offered by their flexibility, low weight of materials, and the cost effectiveness of large-area roll-to-roll (R2R) manufacturing techniques [1]. Despite these advantages, PSCs still suffer from relatively low device performance when compared to inorganic material-based solar cells. In order to overcome low device performance, many research groups are actively studying several critical subjects such as active material development, blend morphology control, interface engineering, and device geometry development [2]. Among these, the development of active material is the most significant challenge for high performance PSCs, especially the development of donor polymers. To achieve a high performance PSC, donor polymers must satisfy several properties: (i) a low band gap for efficient light harvesting; (ii) a low highest occupied molecular orbital (HOMO) level for high open circuit voltage (V_{oc}), as well as adequate energy offset with the lowest unoccupied molecular orbital (LUMO) level of the acceptor; (iii) high charge carrier mobility; and (iv) sufficient solubility in common organic solvents

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http://dx.doi.org/10.1016/j.solmat.2014.02.038 0927-0248/© 2014 Elsevier B.V. All rights reserved. for solution processing [3]. Of various approaches to satisfy these properties of the donor polymer, the most promising one is a "push–pull" or "donor–acceptor" (D–A) concept copolymer, which consists of alternating electron-rich and electron-deficient co-monomers [4]. D–A copolymer can facilitate the tuning of the absorption band as well as efficiently tailor HOMO/LUMO energy levels by combining various types of donors and acceptors [5]. In addition, D–A copolymer can induce strong and efficient charge transport due to strong intermolecular dipole interactions between donor and acceptor moieties [6]. Therefore, a high power conversion efficiency (PCE) of up to 8% has been made possible by development of these D–A copolymers [7].

In addition, in order to further improve the PCE of PSCs, many researchers have introduced various processing techniques such as thermal annealing, solvent annealing, and the inclusion of solvent additives [8]. Recently, among these various processing techniques, the solvent additive method has become more widely adopted because of its simplicity, and, consequently, this approach has led to significant improvements in the PCE. These increases in PCE are attributed to the well-defined BHJ nanomorphology in the active layer, through addition of the solvent additive. However, it is difficult to select a suitable solvent additive for each active material because of each additive's various factors. For example, it has been shown that the active layer nanomorphology and photovoltaic properties of PSCs treated with solvent additive are significantly affected by the type and concentration of the solvent additive [9,10]. Therefore, it is important and necessary to study solvent additive effects in various polymer-based PSCs because it is not fully understood how the type and concentration of the solvent additive affects the photovoltaic performance of polymer:fullerene-based PSCs.

In our previous report, we studied alkyl-substituted thienylenevinylene and phthalimide-based D–A copolymer (PTVPhI-Eh) for the application to ambipolar organic thin film transistor (OTFT) [4](d). This polymer shows an ambipolar behavior in field-effect transitors (μ_{hole} =0.75 cm²/Vs and $\mu_{electron}$ =0.06 cm²/Vs), a band gap of 1.8 eV, and a lower HOMO level of – 5.2 eV when compared to the HOMO level of P3HT (–4.9 eV), as well as high solubility in common organic solvents (~30 mg/mL, chlorobenzene). These properties are well-suited to high performance PSCs, especially the lower HOMO level, which is expected to display a high V_{oc} value of > 0.8 V when used as a donor in polymer:fullerene-based PSCs. In addition, strong absorption in the visible range is expected to be suitable for a front-cell in a series-connected tandem solar cell.

In this paper, we report the photovoltaic properties of a PSC using PTVPhI-Eh polymer as a donor. As expected, PTVPhI-Eh: [6,6]-phenyl- C_{71} -butyric acid methyl ester (P C_{71} BM)-based PSCs exhibited high V_{oc} (0.87 V) because of the deeper HOMO energy level of the PTVPhI-Eh polymer. We also investigate how nanomorphology evolution and photovoltaic properties are affected by blend ratio and various solvent additives such as 1-chloronaphthalene (CN) and 1,8-octanedithiol (ODT). Improved nano-scale phase separated morphology and 4.14% PCE was achieved under conditions of 1:1 wt/wt blend ratio and 2 vol% CN. According to the results of the present study, the photovoltaic properties of PTVPhI-Eh:PC₇₁BM-based PSCs were significantly affected by concentration of the solvent additive. In particular, the V_{oc} showed the most noticeable change caused by the concentration of the solvent additive. Herein, we carry out detailed

analysis of the correlation between solvent additive concentration and V_{oc} in PTVPhI-Eh:PC₇₁BM blend devices using transmission electron microscopy (TEM), X-ray diffraction (XRD), and absorption spectroscopy.

2. Experiments

2.1. Materials

PTVPhI-Eh was synthesized in our laboratory as reported previously [4](d). The number of average molecular weight (M_n) and weight average molecular weight (M_w) were 18,000 g/mol and 36,000 g/mol, respectively [6,6]. Phenyl-C₇₁-butylic acid methyl ester (PC₇₁BM) and poly(3,4-ehtylene dioxythiophene): poly(styrene sulfonate) (PEDOT:PSS, CLEVIOS AI 4083) were purchased from Nano C and H.C.Starck, respectively.

2.1.1. Synthetic procedure of PTVPhI-Eh [4](d)

5,5'-ditrimethylstannyl-(E)-1,2-(3,3'-didodecyl-2,2'-dithienyl) ethylene (0.5 mmol) and N-(2-ethylhexyl)3,6-dibromophthalimide (0.5 mmol) were added together with Pd₂(dba)₃ (0.01 mmol) and p(o-tolyl)₃ (0.04 mmol) to a flask. After the flask was purged with nitrogen, this mixture was dissolved in anhydrous chlorobenzene. The mixture was refluxed at a constant temperature of 120 °C for 48 h. After reaction, the resultant reaction mixture was cooled to room temperature and precipitated from a solution in which 150 mL of methanol was mixed with 30 mL of HCl. The final polymers were purified by multiple Soxhlet extractions with methanol, hexane and chloroform. ¹H NMR (400 MHz, CDCl₃): δ 7.79 (s, 2H), 7.71 (s, 2H), 7.12 (s, 2H), 3.62 (br, 2H), 2.75 (br, 4H), 1.89 (br, 1H), 1.69 (br, 4H), 1.25 (m, 44H), 0.86 (m, 12H). Anal.



Fig. 1. (a) Molecular structures of PTVPhI-Eh and PC₇₁BM. (b) PSC device structure. (c) HOMO/LUMO energy levels of P₃HT, PTVPhI-Eh, and PC₇₁BM. (d) UV-vis absorption spectra of PTVPhI-Eh.

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