

Interface-engineering additives for inverted BHJ polymer solar cells

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ABSTRACT

We report on the photovoltaic (PV) effect of inverted polymer solar cells (PSCs) with a bulk-heterojunction (BHJ) PV layer consisting of a donor-conjugated polymer of poly(3-hexylthiophene) (P3HT) and an acceptor of [6,6]-phenyl C₆₁ butyric acid methyl ester (PCBM) mixed with an interface-engineering additive of poly(oxyethylene tridecyl ether) (PTE). The electron-selective layer used is sol-gel processed zinc oxide (ZnO), and the anode layer is silver (Ag). Improved performance was achieved using the PTE additive in the BHJ PV layer of the inverted PSCs. A 1.77 wt% mixing level of PTE into a blended P3HT:PCBM PV layer with a ratio of 1.00:0.73 produces a power conversion efficiency (PCE) of 3.14%, which was much higher than the reference device (2.77%) without PTE. This improvement can be attributed to not only the improved carrier transport due to the PTEs at the interfaces between the BHJ PV layer and the electrodes, but also the increased carrier lifetime due to the PTE molecules at the interfaces between the phase-separated BHJ domains. These results suggest that the BHJ PV layer mixed with the PTE interface-engineering additive is a promising functional composite material system for use in efficient inverted polymer PV cells.

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1. Introduction

Recently, important research on polymer solar cells (PSCs) has generated intense interest since photo-induced electron transfer from an electron donor conjugated polymer to an acceptor fullerene was reported [1–5]. An interesting study involved the efficient photo-induced charge generation of a photovoltaic (PV) layer of bulk heterojunction (BHJ) PSC [2,3], which is composed of interpenetrating channel-like domains of the separated polymer and fullerene phases in the blended PV layer. After annealing at elevated temperatures together with the BHJ structure, efficient PSCs with a power conversion efficiency (PCE) of 3–5 % have been demonstrated using PV layers of poly(3-hexylthiophene) (P3HT) and phenyl C₆₁-butyric acid methyl ester (PCBM). The annealed BHJ films fabricated with these materials show well-defined bicontinuous interpenetrating networks [4,5]. However, although the P3HT:PCBM BHJ PV layer between a transparent indium-tin-oxide (ITO) anode and a low work-function Al cathode has been successful in reaching efficiencies of about 5%, new device structures are needed to further improve not only efficiencies but also stability, because the use of regular forward device structure in conventional PSCs is limited due to poor stability of the Al cathode [6]. Instead of Al, by applying a stable cathode material to PSCs, it is possible to increase the device lifetime [6].

One of the most stable and efficient electron transporting materials to date is zinc oxide (ZnO), due to its high electron mobility [7–9]. Based on ZnO electron-selective layers, inverted BHJ PSCs, in which the charge-collecting nature of the electrodes is reversed, have been developed. In these devices, a modified ITO is used as the cathode, and a high-work-function metal is used as the anode such as (ITO cathode/ZnO electron-selective layer/P3HT:PCBM BHJ PV layer/MoO₃ hole-selective layer/Ag anode). PCEs up to 3% have been reported for inverted BHJ PSCs [7–9]. Recently, a PCE of 5–6 % was reported for an inverted BHJ PSC based on a blend of the phenyl C₇₁-butyric acid methyl ester and low-band-gap polymer of poly[N-9''-hepta-decanyl-2,7-carbazole-alt-5,5-(4',7'-di-2-thienyl-2',1',3'-benzothiadiazole)] [10], demonstrating the best performance of any single junction inverted BHJ PSC studied to date.

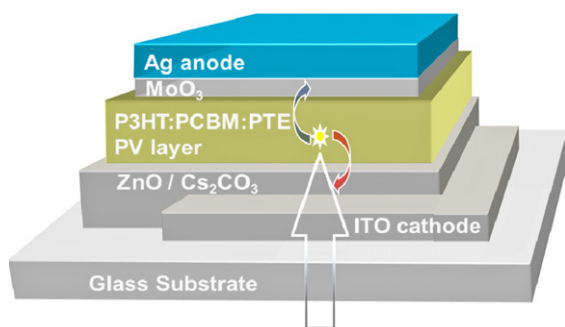
More recently, it was also suggested that by incorporating a few volume percent of nonionic poly(oxyethylene tridecyl ether) (PTE) surfactant as additives in the BHJ PV films of the conventional forward PSCs in view of its low highest-occupied-molecular-orbital (HOMO; –8.1 eV) and high lowest-unoccupied-molecular-orbital (LUMO; –2.1 eV) [11]. The PCE could be increased by improving the dissociation efficiency and carrier transport at the interfaces between the phase-separated donor polymer and fullerene domains (BHJ interfaces), and between the PV layer and the cathodes [12,13]. In these BHJ PV layers, the surfactants in the PV layers functioned as “interface engineering additives” [13] to reduce recombination loss *via* preserving separated charge carriers or increasing the carrier transport at the interfaces. Thereafter,

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several efforts have been explored to improve the device performance of BHJ PSCs with surfactants, and several studies have reported highly efficient PSCs fabricated using the interface-engineering additives. However, in contrast to the BHJ interfaces and the interfaces between the PV layer and the cathode (cathode interface), relatively few studies have been conducted on the effects of the PTE additive at the interface between the PV layer and the anode (anode interface) on PV performance. In order to improve the degree of dissociation and carrier lifetime in the BHJ PSCs, further research is required for these interfaces. Moreover, no study has been conducted on interface-engineered inverted BHJ PSCs with the additives until now.

In this study, to understand the effects of PTE additive on the BHJ interfaces and the anode interface on PV performance, we investigated inverted BHJ PSCs with a blended P3HT:PCBM PV layer mixed with a PTE surfactant additive (Fig. 1) and compared the performance with inverted BHJ PSCs without the PTE. In the inverted BHJ PSCs with PTE, it is expected that the PTE molecules at BHJ interfaces may effectively block holes from the donor P3HT polymer domains and draw electrons to PCBM domains, thereby improving the selective collection charges at the BHJ interfaces. From the experiments, it was successfully demonstrated that more highly efficient inverted BHJ PSCs are realized by introducing PTE surfactant into the P3HT:PCBM layer. The PCE increased from 2.77% to 3.14% via increase in the carrier lifetime for a ~ 1.77 wt% mixing level of the PTE surfactant into the P3HT:PCBM (1.00:0.73) BHJ PV layer.

Inverted BHJ PSC structure



Forward stacked PSC structure

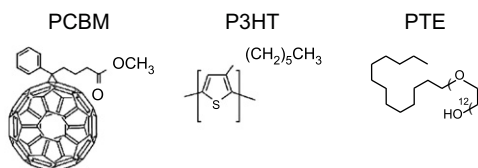
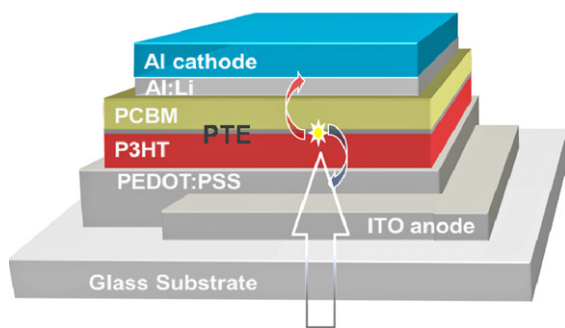


Fig. 1. Device and chemical structures used of the inverted BHJ PSCs and the model devices of the forward stacked PSCs investigated. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

2. Experimental details

2.1. Fabrication of inverted BHJ polymer solar cells

P3HT (Rieke Metals, Inc.), PCBM (Nanostructured Carbon Inc.), PTE ($C_{13}H_{22}(OCH_2CH_2)_{12}OH$, Aldrich), MoO_3 (Sigma–Aldrich), and poly(3,4-ethylenedioxythiophene):poly(4-styrenesulphonate) (PEDOT:PSS, Clevis PVP. Al 4083, H.C. Starck Inc.) were used as received from the manufacturers. The structures of the materials used in the photoactive layer are shown in Fig. 1.

The inverted BHJ PSCs were fabricated on glass using an inverted sandwich-like configuration, with an ITO (80 nm, $30 \Omega/\text{square}$) cathode modified by an electron-collective ZnO interlayer and a silver (Ag) anode. Inverted BHJ PSCs were built according to the following procedure: ITO-coated glass substrates were cleaned by ultrasonic treatments in acetone and isopropyl alcohol. For an electron-selective ZnO layer, a sol-gel derived ZnO precursor was spin-coated onto the ITO surface after UV-ozone treatment for 5 min. The details of the preparation method are similar to those described previously [14]. As a zinc source, zinc acetate-2-hydrate [$Zn(CH_3COO)_2 \cdot 2H_2O$] was utilized. The solvent and stabilizer used were 2-methoxyethanol ($CH_3OCH_2CH_2OH$) and ethanolamine, respectively. The concentration of zinc acetate was chosen to be 5 wt%, and the precursor solution was mixed thoroughly by a magnetic stirrer at $60^\circ C$ for 2 h. The ZnO precursor was spin-coated on top of the ITO glass at 1000 rpm for 100 s, and subsequently pre-heated at a temperature of $300^\circ C$ for 10 min to evaporate the solvent and remove organic residuals. The process was repeated four times to obtain a desired thickness (*ca.* 135 nm). After the deposition of the last layer, the films were inserted into a furnace and annealed in ambient atmosphere at the temperature of $450^\circ C$ for 60 min to crystallize the ZnO thin film. X-ray diffraction was performed on the sol-gel processed ZnO films. The annealed ZnO films show two strong crystalline ZnO peaks at 30.3° (100) and 34.3° (002). A thin cesium carbonate (Cs_2CO_3) electron-collecting buffer layer (~ 5 nm) was also spin-coated on top of the ZnO layer [15]. Next, for preparing the active BHJ PV layer, P3HT (2.40 wt%), PCBM (1.76 wt%), and PTE additive (0.075 wt%) were dissolved in 1,2-dichlorobenzene at $40^\circ C$ for 24 h. The resulting solution was spin-coated on top of the Cs_2CO_3 buffer layer at 1200 rpm for 90 s and then annealed at $120^\circ C$ for 3 min in order to fabricate the P3HT:PCBM:PTE BHJ PV layer. The BHJ PV layer had a thickness of about 220 nm. On top of the active BHJ PV layer, for the hole-collecting interlayer, a *ca.* 20 nm-thick MoO_3 interlayer was formed by thermal deposition (0.05 nm/s) at a base pressure below 2.5×10^{-4} Pa. A pure Ag (*ca.* 100 nm thick) anode layer was then formed on the MoO_3 interlayer under the same vacuum conditions. Finally, after the inverted BHJ PSCs had been fabricated, the devices were annealed at $150^\circ C$ for 10 min in a N_2 -filled glove box to induce the crystallization of the PV layer (*i.e.*, post-thermal annealing) [4,5]. Thus, the device structure consisted of the sequence, [ITO cathode/ZnO/ Cs_2CO_3 /P3HT:PCBM:PTE/ MoO_3 /Ag anode] (sample cell). The active area of the fabricated device was $3 \times 3 \text{ mm}^2$. For comparative purposes, we also fabricated a reference PSC using the sequence [ITO cathode/ZnO/ Cs_2CO_3 /P3HT:PCBM/ MoO_3 /Ag anode] without PTE additive (reference 1). We also fabricated another reference PSC that had a PTE interfacial layer (*ca.* 1–2 nm) on the PV layer, *i.e.*, [ITO cathode/ZnO/ Cs_2CO_3 /P3HT:PCBM/PTE/ MoO_3 /Ag anode] (reference 2), by spin-coating a PTE solution (0.075 wt% in distilled water) on the P3HT:PCBM layer. We monitored the thickness of the PTE interfacial layer using ellipsometry. It should be noted that apart from the differences in the PV layers as described, all the reference PSCs were fabricated using exactly the same process as that used for the sample cell.

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