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Successful incorporation of optical spacer and additive solvent for enhancing the photocurrent of polymer solar cell

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ABSTRACT

An optical spacer and a processing additive (PA) offer various advantages in the enhancement of the photocurrent in a polymer solar cell, but successful incorporation of both in one device has yet to be demonstrated. Indeed, an optical spacer is considered to conflict the use of a PA in the solar cell system. Here we demonstrate a significant enhancement of the photocurrent in a polymer solar cell via successful incorporation of a solution-processable titanium sub-oxide (TiO_x) optical spacer and a PA. By using a simple, non-material-specific, and effective vacuum treatment before deposition of the TiO_x optical spacer, the conflict between the optical spacer and PA was resolved and remarkable enhancement in photocurrent was attained without any loss in the fill factor and open-circuit voltage. When the TiO_x optical spacer was introduced together with the 1,8-diiodoctane additive, the power-conversion efficiencies were significantly improved from 7.63% to 9.33% for the PBDTTT-EFT:PC₇₀BM device and from 5.5% to 6.46% for the P(BDT-TDPPDT-TPD)/PC₇₀BM device, where PBDTTT-EFT stands for poly[4,8-bis(5-(2-ethylhexyl)thiophen-2-yl)benzo[1,2-*b*;4,5-*b'*]dithiophene-2,6-diyl-alt-(4-(2-ethylhexyl)-3-fluorothieno[3,4-*b*]thiophene-)-2-carboxylate-2-6-diyl], PC₇₀BM stands for [6,6]-phenyl-C₇₁-butyric acid methyl, and P(BDT-TDPPDT-TPD) stands for poly[benzodithiophene-4,6-*bis*(thiophen-2-yl)-2,5-dioctylpyrrolo[3,4-*c*]pyrrole-1,3(2H,5H)-dione-5-octylthieno[3,4-*c*]pyrrole-4,6-dione].

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1. Introduction

Polymer solar cells (PSCs) are a promising means to provide a low-cost, wearable, printable, and flexible source of energy [1–5]. Although PSCs are now very close to commercialization and their power-conversion efficiency (PCE) is approaching 11%, further improvements in efficiency are still required to reduce the cost of electricity in terms of dollar per kilowatt-hour [6–18]. Enhancing the photocurrent of the PSC is a reasonable strategy for improving the PCE because it can be easily obtained by increasing photon absorption in the bulk heterojunction (BHJ) active layer or improving the internal quantum

efficiency (IQE) of the device, or a combination of enhancing both the photon absorption and IQE [5–7,11–15].

The use of an optical spacer in the PSC is a feasible route to enhance the photocurrent [5,19,20]. The optical spacer redistributes light intensity within the BHJ by changing the optical interference between the incident light and light reflected from the metal cathode, leading to better harvest of light in the BHJ layer [5,19,20]. In addition, it can be achieved without increasing the active-layer thickness, thus avoiding an increase in the internal resistance of the device. The solution-processable titanium sub-oxide (TiO_x) composite is widely used as an optical spacer owing to its superior properties such as high transparency in the visible range, well-matched energy level, and high electron mobility. Because of the high energy level of its highest occupied molecular orbital (HOMO), TiO_x can also serve as a hole blocker to enhance the performance of the PSC with higher fill factor (FF) [5,19,21,22].

Improving the IQE is another strategy to enhance the photocurrent [5,23,24]. In the BHJ polymer blend, since generated

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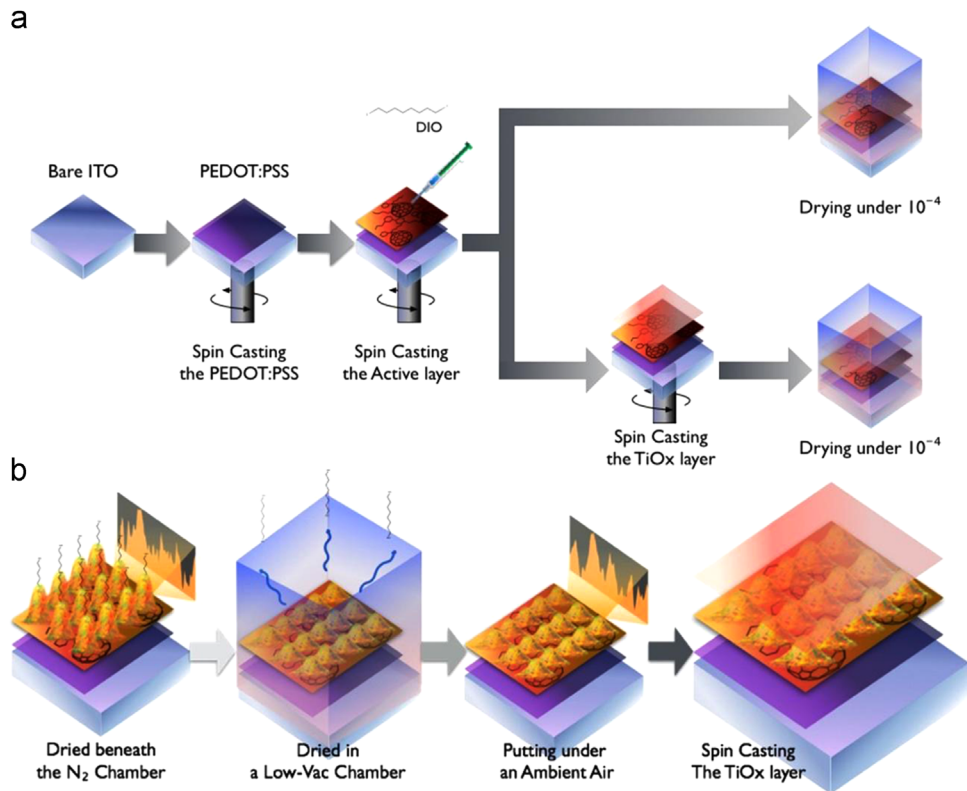


Fig. 1. (a) Schematic diagram of the fabrication processes of PSCs with and without the TiO_x layer. (b) Diagram of process for preparing the BHJ/TiO_x film using low-vacuum treatment.

excitons are separated at the interface between the polymer and the fullerene derivative, and the separated charges are transported to the electrodes via self-assembled bicontinuous charge-transport channels, optimizing the morphology of the BHJ provides the route to high IQE [25]. It has been proved that the processing-additive (PA) technique is the most successful way to optimize the BHJ morphology and achieve better phase separation on the nanometer scale [26–30]. Adding a small amount of additive with selective solubility and high boiling point to the BHJ solution prevents the formation of large clusters of fullerene derivatives and provides better nanometer-scale morphology of the BHJ, leading to higher IQE [26–30].

Despite such beneficial advantages of the TiO_x optical spacer and the PA, successful incorporation of the TiO_x layer with active layers processed with the PA has not been demonstrated so far. Indeed, using TiO_x as an optical spacer conflicts with the PA technique in the PSC [30,31]. For example, Lee et al. reported that including a small amount of 1,8-diiodooctane (DIO) as an additive resulted in uneven topography of the BHJ layer and plummeted the effect of the optical spacer owing to the destructive interference between the BHJ and TiO_x layers. The inhomogeneous interface of the two layers gave rise to a large decrease in the FF of the device [30]. More recently, Kwon et al. demonstrated the positive synergistic effect of the PA and optical spacer by choosing 1-chloronaphthalene (1-CN) instead of DIO as the additive. They emphasized that the addition of 1-CN resulted in smoother and more desirable nanoscale BHJ morphology, and the TiO_x optical spacer enhanced the device performance [31]. Although they succeeded in increasing the PCE via the synergy between the TiO_x optical spacer and the PA, its use was restricted to the PSC process with 1-CN additive. Moreover, the PSC continued to exhibit obviously reduced FF.

Herein we report that we successfully incorporated a TiO_x optical spacer together with a PA to enhance the photocurrent of the PSC. By

introducing a low-vacuum (LV) treatment before deposition of the TiO_x optical spacer, we obtained remarkable enhancement in the photocurrent without any loss in FF and the open-circuit voltage (V_{oc}). The PCE increased from 5.5% to 6.46% for the ITO/PEDOT:PSS/P(BDT-TDPPDT-TPD):PC₇₀BM/TiO_x/Al device [32] and from 7.63% to 9.33% for the ITO/PEDOT:PSS/PBDTTT-EFT:PC₇₀BM/TiO_x/Al device [29,33], where PEDOT:PSS is the abbreviation of poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate), P(BDT-TDPPDT-TPD) stands for poly [benzodithiophene-4,6-bis(thiophen-2-yl)-2,5-dioctylpyrrolo[3,4-c]pyrrole-1,3(2H,5H)-dione-5-octylthieno[3,4-c]pyrrole-4,6-dione], PC₇₀BM stands for [6, 6]-phenyl-C₇₁-butyric acid methyl, and PBDTTT-EFT stands for poly[4,8-bis(5-(2-ethylhexyl)thiophen-2-yl)benzo[1,2-*b*:4,5-*b'*]dithiophene-2,6-diyl-alt-(4-(2-ethylhexyl)-3-fluorothieno[3,4-*b*]thiophene-)-2-carboxylate-2-6-diyl)].

2. Experimental section

The Fourier-transform infrared (FTIR) spectroscopy data were obtained with a PerkinElmer Spectrum GX FT-IR spectrometer. The BHJ films with and without DIO were fabricated on potassium bromide (KBr) substrates for the FTIR measurements. For the films with DIO, one film was left in a glove box under atmospheric pressure and another film was placed in a dry oven under a nitrogen atmosphere at $\sim 10^{-2}$ Pa for 10 min.

The PSCs were fabricated with the simple device structure of (ITO-coated glass substrate)/PEDOT:PSS/polymer:PC₇₀BM/Al. The ITO-coated glass substrate was first cleaned with detergent, ultrasonicated in acetone and isopropyl alcohol, and then dried overnight in an oven. PEDOT:PSS (Baytron PH) was spin-cast from an aqueous solution, after which the substrate was dried for 10 min at 140 °C in air and then transferred to a glove box to spin-cast the photoactive layer. A solution containing a mixture of P(BDT-TDPPDT-TPD)-PC₇₀BM (1:1.5 wt%) in chlorobenzene (CB)

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