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## Ternary polymer solar cell based on two donors and one acceptor for improving morphology and power conversion efficiency



SYNTHETIC METAL

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#### A R T I C L E I N E O

#### A B S T R A C T

of the thin film, respectively.

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#### 1. Introduction

In order to achieve high power conversion efficiency (PCE) in bulk heterojunction (BHJ) polymer solar cells (PSCs), various methods for improving the open circuit voltage  $(V_{oc})$  [\[1,2\],](#page--1-0) fill factor (FF) [\[3\]](#page--1-0), and short circuit current  $(J_{\rm sc})$  have been suggested [\[4,5\]](#page--1-0).  $V_{\text{oc}}$  is determined by the intrinsic properties of the donor and acceptor materials depending on their molecular structure, whereas  $J_{\rm sc}$  can be affected by the charge carrier mobility and internal morphology organized with the donor and acceptor materials.

To improve the  $J_{\rm sc}$  value in a BHJ PSC, p-type and n-type materials should be well mixed, maintaining nanoscopic crystalline domains whose dimensions are smaller than the optimized exciton diffusion length  $(\sim10 \text{ nm})$ . Nanophase separation in blend films is especially important for maximizing the interfacial contact area that facilitates interconnections between the crystalline domains for effective charge transport [\[6,7\]](#page--1-0).

Recently, modification method of internal morphology by combining various materials has been proposed as a way to induce change to the morphology  $[8,9]$ . Attention has been paid to optimize ternary PSC systems for facilitating charge transport without undesired energy transfer and for extending the light

<http://dx.doi.org/10.1016/j.synthmet.2016.07.009> 0379-6779/@ 2016 Elsevier B.V. All rights reserved. harvesting wavelength to the solar emission region. Selection of appropriate materials and formation of a well-miscible ternary morphology are important because it is generally difficult to control the miscibility of three components (two different polymers and a fullerene derivative) and the individual crystallization of each component [10–[13\]](#page--1-0).

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We synthesized isoindigo-based two donor-acceptor (D-A) copolymers to optimize the morphology in ternary blend films with two p-type polymers and n-type phenyl-C61-butyric acid methyl ester  $(PC_{61}BM)$ . Compared to binary systems with a single copolymer and  $PC_{61}BM$ , the ternary polymer solar cell with two copolymers and  $PC_{61}$ BM showed highly enhanced power conversion efficiency of 5.13% with a high short circuit current of 13.23 mA cm<sup>-2</sup>. These results were supported by atomic force microscopy (AFM) and transmission electron microscopy (TEM) observations of the surface and internal morphology

> Thus, various attempts have been made to improve the desired morphology in ternary PSC system. Among them, control of the donor [\[14\]](#page--1-0) or acceptor [\[15\]](#page--1-0) mixing ratios for nanoscale phase separation is a useful strategy. The well-controlled ternary morphology can lead to the formation of smaller intermixed crystalline domains and favorable molecular orientation, which in turn could be beneficial to enhance the short circuit current and PCE.

In this study, two different isoindigo-based donor-acceptor (D-A) copolymers were applied to the ternary blend system for PSCs. Since isoindigo-based polymers have been reported to have broad absorption and deep highest occupied molecular orbital (HOMO) energy levels, they were chosen as the active components for preparing the active layer [\[16,17\].](#page--1-0) For most of isoindigo-based polymers, the polymer backbones were designed to have structural similarity, in order to improve the miscibility between two polymers that would be mixed together in the ternary blend system. Therefore, isoindigo-based polymers were synthesized using 1,2-bis(5-(trimethylstannyl)-thiophen-2-yl)ethene (IIDT) [\[18\]](#page--1-0) and 1,2-bis(5-(trimethylstannyl)-selenophen-2-yl)ethene Corresponding authors.  $(IIDS) [19]$  $(IIDS) [19]$  as the structurally similar donor moieties.



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A random terpolymer containing two different donors, 1,2-bis (5-(trimethylstannyl)-thiophen-2-yl)ethene and 1,2-bis(5-(trimethylstannyl)-selenophen-2-yl)ethene, was also synthesized as the control polymer. The  $(IIIDT:IIDS = 1:1): PC_{61}BM = 1:1.5$  ternary system showed highly favorable internal morphology, and the corresponding BHJ PSC showed a relatively high  $J_{\rm sc}$  of 13.23 mA  $cm^{-2}$  and PCE of 5.13% compared to those of the binary system (IIDT:PC $_{61}$ BM and IIDS:PC $_{61}$ BM) in BHJ PSCs. These results were supported by analysis of the internal morphology in film states using atomic force microscope (AFM) and transmission electron microscopy (TEM).

#### 2. Experimental section

#### 2.1. Materials and synthesis

All reagents for preparing the three polymers were purchased from Sigma-Aldrich, TCI, and Acros Organics. Reagent-grade solvents were purified by a solvent dispensing system (J. C. Meyer, Laguna Beach, CA, USA). IIDT and IIDS were synthesized following the literature methods [\[18,19\].](#page--1-0)

#### 2.2. Synthesis of IIDTS

(E)-6,6′-Dibromo-1,1′-bis(2-decyltetradecyl)-[3,3′-biindolinylidene]-2,2-dione (150 mg, 0.14 mmol), 1,2-bis(5-(trimethylstannyl)-thiophen-2-yl)ethene (35.5 mg, 0.07 mmol) and 1,2-bis (5-(trimethylstannyl)-selenophen-2-yl)ethene (41.9 mg, 0.07 mmol) were dissolved in dry toluene (10 mL) and the solution was degassed with nitrogen for 0.5 h. Tris(dibenzylideneacetone) dipalladium (0) (2.5 mg, 2 mol%) and tri-o-tolylphosphine (3.3 mg, 8 mol%) were then added to the mixture at 100 $\degree$ C and the reaction was kept for 6 h. Then, the reaction mixture was cooled to room temperature and added dropwise into methanol (200 mL). The crude polymer was collected by filtration and dried at  $40^{\circ}$ C under vacuum for 2 h. The pre-dried polymer was then purified by Soxhlet extraction with methanol, acetone, hexane, and chloroform, sequentially. IIDTS was obtained by precipitating the chloroform fraction in methanol and the solid product was dried under vacuum for 24 h. Yield: 80% (130 mg).  $(M_n = 23.1 \text{ kDa})$ PDI = 3.56). Element Anal. Cal: C, 75.97; H, 9.61; N, 2.36; S, 2.70. Found: C, 75.90; H, 9.49; N, 2.39; S, 2.74. (m:n = 1:1).

#### 2.3. Instrumentation

Elemental analysis was performed using a Thermo Scientific Flash 2000 (Thermo Fisher Scientific) elemental analyzer to determine the contents of C, H, N, and S. Average molecular weights (MWs) of the polymers were determined by gel permeation chromatography (GPC, Waters GPC, Waters 515 pump, Waters 410 RI, 2 x PLgel Mixed-B) using a polystyrene standard in 1.2-dichlorobenzene ( $o$ -DCB) as the eluent at 80 $\degree$ C.

Absorption spectra for both films and solutions were recorded using a UV–vis absorption spectrophotometer (HP 8453, photodiode array) over the wavelength range of 190–1100 nm. A chloroform solution of the polymers was filtered through an Acrodisc syringe filter (Millipore  $0.45 \,\mathrm{\upmu m}$ ), and the thin film was prepared by spin-coating on a quartz glass substrate.

Electrochemical properties were evaluated by cyclic voltammetry (CV) using an eDAQ EA161 potentiostat. The thin film was coated on a platinum plate using a polymer solution in chloroform, while 0.10 M tetrabutylammonium hexafluorophosphate  $(Bu_4NPF_6)$  in anhydrous acetonitrile was selected as the electrolyte. Ag/AgCl and platinum wire were used the reference and counter electrodes, respectively.

Atomic force microscopy (AFM; Advanced Scanning Probe Microscope, XE-100, PSIA) was employed to investigate the surface morphology of polymer films. A silicon cantilever was used in tapping mode for all scans.

All transmission electron microscopy (TEM) images of the blend films with polymers and  $PC_{71}$ BM were recorded with on Tecnai G2 F30 transmission electron microscope (FEI Inc.; operating voltage, 300 kV). The blend films were deposited on the surface of PEDOT: PSS coated on ITO. The blend films were immersed in deionized water (5 min) and floated onto the air/water interface. The pieces of the film were picked up and placed on carbon-coated copper grids for recording the images.

#### 2.4. Fabrication and characterization of PSCs

The patterned indium tin oxide (ITO) glass was sequentially cleaned in acetone, deionized water, and isopropyl alcohol by sonication, then dried in a vacuum oven for 10 h at 120 $\degree$ C and treated with  $O_2$ -plasma (CR403 M, 90 W) for 20 min. A thin layer (40 nm) of PEDOT:PSS was spin-coated on the ITO glass.



Fig. 1. Synthesis of IID-based copolymers and random terpolymer.

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