



Efficient ternary polymer solar cells with a shelf-life stability for longer than 410 days

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ABSTRACT

We have prepared an efficient ternary polymer solar cell incorporating a poly(indacenodithiophene)-based conjugated polymer (PIDTBT), [6,6]-phenyl-C₇₁-butyric acid methyl ester (PC₇₁BM), and 3,9-bis(2-methylene-(3-(1,1-dicyanomethylene)indanone)-5,5,11,11-tetrakis(4-hexylphenyl)-dithieno[2,3-*d*:2',3'-*d'*]-s-indaceno[1,2-*b*:5,6-*b'*]-dithiophene (ITIC). Grazing-incidence wide-angle X-ray scattering (GIWAXS), atomic force microscopy (AFM), and photoluminescence (PL) spectroscopy revealed that intermolecular interactions between the PC₇₁BM and ITIC units disrupted the formation of large ITIC crystals. The ITIC and PC₇₁BM components formed compatible domains that dispersed well within the PIDTBT matrix, providing optimized ternary blends for efficient carrier transport and led to greater photon-to-electron conversion efficiency. Compared with the pre-optimized PC₇₁BM binary device, the ternary device displayed an improvement in short circuit current density (J_{sc}) from 12.0 ± 0.3 to 14.2 ± 0.5 mA cm⁻², due primarily to complementary light harvesting in the visible and near-infrared regions; as a result, the device performance improved by 20%—from 5.5 ± 0.2 to $6.6 \pm 0.1\%$ under AM 1.5G (100 mW cm⁻²) irradiation. Furthermore, the ternary cell exhibited outstanding long-term stability, with its performance remaining high (at 6.8%) after storage for 410 days in a glove box (ISOS-D-1 (shelf life-time)).

1. Introduction

Polymer solar cells (PSCs) have high flexibility and low weight, can be prepared using high-throughput roll-to-roll manufacturing, are simple to install, and have proven durability; accordingly, they are recognized as one of the most promising next-generation energy sources [1–8]. Progress in the development of n- and p-type materials, as well as advance, in device engineering, have led to the power conversion efficiencies (PCEs) of PSCs surpassing the 10% (certified) milestone, with the highest reported PCE reaching 13% [9–14]. Nevertheless, the conjugated polymers used in PSCs typically display low carrier mobility (relative to those of inorganic semiconductors) and, thus, the thickness of the active layer (ca. 100 nm) is limited to achieve balanced carrier collection and transport. Furthermore, the narrow intrinsic absorption and limited open-circuit voltage (V_{oc}) of single P-type materials [the value of V_{oc} is governed by the difference between the energy level of the highest occupied molecular orbital (HOMO) of the P-type material (Donor_{HOMO}) and the lowest unoccupied molecular orbital (LUMO) of the N-type material (Acceptor_{LUMO}); low-band-gap materials have high-energy HOMOs and are, therefore, associated with low values of V_{oc}] mean that full harvesting of the solar spectrum in blends with single-

acceptor materials (most commonly, fullerenes) remains challenging.

Recent developments in novel high-performance non-fullerene N-type materials, conjugated polymers, and the physics of device fabrication have led to ternary PSCs emerging as the major strategy toward enhancing the performance of PSCs [15–19]. In contrast to tandem cells, blending a third component (donor or acceptor) having complementary light-harvesting ability is a simple method of fabrication, which can readily be scaled up, for enhancing device performance. In the main, the most favorable ternary PSCs feature one of two structures: donor 1:donor 2:acceptor (D:D:A) or donor:acceptor 1:acceptor 2 (D:A:A) [17,20]. In some cases, the third component forms a favorable energetic structure or plays the role of improving the morphology of the medium to restrict carrier recombination and improve charge transport [21,22]. Thus, the ternary strategy has great potential for simultaneously improving the light-harvesting ability, carrier transport, and morphology of PSCs, thereby improving their fill factors (FFs) and short-circuit current densities (J_{sc}) [23]. By combining the benzodithiophene (BDT)-based conjugated polymer PTB7-Th and the small molecule DR3TSBDT with PC₇₁BM, Yang and coworkers reported a record-PCE of 12.1% for a ternary PSC featuring a well-controlled molecular orientation [21].

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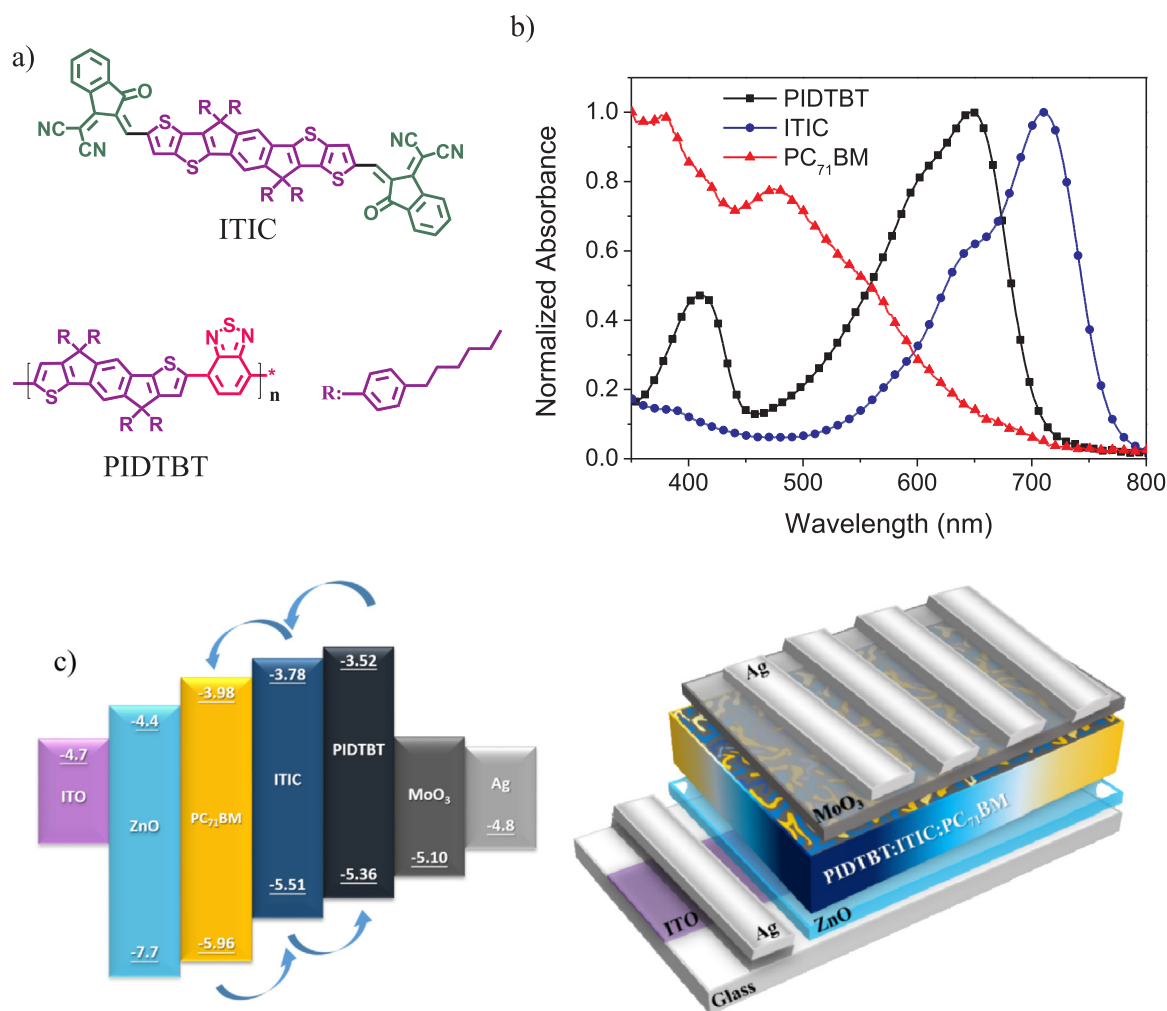


Fig. 1. (a) Chemical structures of PIDTBT and ITIC. (b) UV-Vis spectra of thin films of PIDTBT, PC₇₁BM, and ITIC. (c) Energy diagrams of the materials and device architecture used in this study.

The indacenodithiophene (IDT)-based conjugated polymer PIDTBT had provided devices displaying promising values of V_{oc} (> 0.85 V) and PCE (ca. 6%) [24,25]. Nevertheless, because of the intrinsic absorption behavior of PIDTBT, the absence of light harvesting in the near-infrared (NIR) region has weakened the PSC performance of PIDTBT-based binary devices. Recently, Zhan et al. reported an indacenodithieno[3,2-*b*]thiophene (IT)-based material, [3,9-bis(2-methylene-(3-(1,1-dicyanomethylene)-indanone)-5,5,11,11-tetrakis(4-hexylphenyl)-dithieno[2,3-*d*:2',3'-*d'*]-*s*-indaceno[1,2-*b*:5,6-*b'*]-dithiophene (ITIC); Fig. 1a], having its onset absorption at a wavelength of 800 nm, as a potential NIR acceptor [26–28]. In an attempt to maintain a high value of V_{oc} while ensuring superior light harvesting, we initially introduced ITIC as a third component within the PIDTBT:PC₇₁BM blend. The relationship between ITIC and BDT-based polymers has been explored extensively, delivering a PCE of greater than 10% [21,29,30]. Nevertheless, studies of IDT-based conjugated polymers and their ITIC derivatives are relatively rare. Fig. 1b reveals that ITIC and PC₇₁BM absorb in a complementary manner in the NIR and low-wavelength regions, respectively. Because PIDTBT provides complementary absorbance at wavelengths between 500 and 700 nm, these three materials might form a suitable ternary composition. Moreover, these materials can form a cascade energetic structure for efficient charge dissociation and transport (Fig. 1c) [30]. Because the energy levels of the HOMO and LUMO of ITIC are located between the HOMO and LUMO energy levels of PIDTBT and PC₇₁BM, efficient exciton dissociation should occur at the donor-acceptor interfaces. The deep HOMO energy level of

PIDTBT should induce a high value of V_{oc} , while the broad light harvesting ability of this ternary system should provide a high value of J_{sc} , together leading to an improved PCE.

We employed tapping-mode atomic force microscopy (AFM), photoluminescence (PL) spectroscopy, UV-Vis spectroscopy, space-charge limited current density (SCLC) measurements, and grazing-incidence wide-angle X-ray scattering (GIWAXS) to examine the effect of the blend ratio of PIDTBT, PC₇₁BM, and ITIC on the optoelectronic properties and device performance. Because of efficient light harvesting and the emergence of a well-defined ternary blend morphology, we observed an enhancement in the value of J_{sc} (from 12.0 ± 0.3 to 14.2 ± 0.5 mA cm⁻²) and a 20% increase in device performance (from $5.5 \pm 0.2\%$ to $6.6 \pm 0.1\%$) for the optimized ternary devices, relative to those of pre-optimized control PIDTBT:PC₇₁BM binary devices. Furthermore, the optimized ternary device exhibited extremely high stability after storage (> 410 days) in a glove box (N₂), with constantly high PCEs of 6.8%.

2. Results and discussion

Careful design and judicious choice of ternary components—that match and can form a cascade of energy levels—are required to ensure high device performance. In this study, ternary PSCs were prepared using materials with compatible energy levels, namely PIDTBT, ITIC, and PC₇₁BM. Fig. 1a displays the molecular structures of PIDTBT and ITIC. Fig. 1b reveals that the absorption spectrum of PC₇₁BM covers

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