



# High efficient ternary polymer solar cells based on absorption complementary materials as electron donor



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## ARTICLE INFO

### Article history:

Received 3 April 2015

Received in revised form

12 May 2015

Accepted 24 May 2015

Available online 11 June 2015

### Keywords:

Ternary polymer solar cells

Absorption spectral complementary

Power conversion efficiency

## ABSTRACT

A series of polymer solar cells (PSCs) were fabricated with narrow band gap polymer poly[[4,9-dihydro-4,4,9,9-tetra(4-hexylbenzyl)-s-indaceno[1,2-b:5,6-b0]-dithiophene-2,7-diyl]-alt-[2,3-bis(3-(octyloxy)phenyl)-2,3-dihydro-quinoxaline-2,20-diyl] (PIDTDTQx), small molecule material 2,4-bis[4-(N,N-diisobutylamino)-2,6-dihydroxyphenyl]squaraine (DIB-SQ), or their blend as electron donor, and [6,6]-phenyl-C71-butyric acid methyl ester (PC<sub>71</sub>BM) as electron acceptor. The champion power conversion efficiency (PCE) of ternary PSCs arrives to 6.49% with a short circuit current density ( $J_{sc}$ ) of 11.56 mA/cm<sup>2</sup> and a fill factor (FF) of 66% when the DIB-SQ doping ratio in donors is 9 wt%. The champion PCE values of binary PSCs with PIDTDTQx:PC<sub>71</sub>BM or DIB-SQ:PC<sub>71</sub>BM as the active layers are 5.47% or 1.78%, respectively. An apparent PCE improvement of 18.6% was obtained from the optimized ternary PSCs compared with binary PSCs with PIDTDTQx:PC<sub>71</sub>BM as the active layers. The underlying reason of PCE improvement was investigated from the absorption spectral complementary, photoluminescence emission quenching, intermolecular charge transfer, and the balance of charge carrier transport in ternary active layers.

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## 1. Introduction

In the past years, the power conversion efficiency (PCE) of polymer solar cells (PSCs) has been greatly improved due to the rapid development of high efficient narrow band gap photovoltaic materials. It is known that PSCs have been considered as one of the most promising candidates to replace the traditional energy source due to their low cost, friendly environment, flexibility and ease of large-area processing [1–4]. For commercial viability of PSCs towards real solar cell applications, solar parks based on PSCs have been built up for analyzing performance, practicality, installation speed, and energy payback time [5,6]. Krebs's group reported some landmarked research results on the fabrication, encapsulation, installment and lifetime of large scale PSCs in advance [7–9]. The bulk heterojunction sandwiched by electrodes is commonly considered as an optimized structure due to the large interface area between donor and acceptor materials that ensures efficient photogenerated exciton dissociation into free charge carriers. Some smart strategies were carried out to adjust donor/

acceptor phase separation to form a bicontinuous interpenetrating network, such as annealing treatment, solvent treatment, solvent additive and mixed solvent [10–13]. To further increase the performance of PSCs, tandem configuration PSCs were designed with different band gap materials as the electron donors for the top and bottom sub-cells [14–18]. It is commonly reported that series connected tandem PSCs exhibit a high open circuit voltage ( $V_{oc}$ ), which is nearly ideal to the summation of the  $V_{oc}$ 's for top and bottom sub-cells, while the  $J_{sc}$  is slightly lower than that of sub-cells. The middle connection layer between top and bottom sub-cells is one of the most important features to obtain high performance tandem PSCs. The middle connection layer should have rather low resistance and high light transmittance, which is a great challenge for the potential application of tandem PSCs with high yield. Recently, we reported a simultaneous improvement in  $J_{sc}$ ,  $V_{oc}$  and fill factor (FF) of PSCs through ternary strategy, leading to a 21.2% PCE improvement compared with binary PSCs [19]. The active layers of ternary PSCs contain three kinds of photovoltaic materials, such as two donors and one acceptor, one donor and two acceptors [20–23]. Ternary PSCs adopt the efficient single bulk heterojunction structure, having the advantage of simple fabrication technology. To intuitively exhibit the PCE improvement based on the tandem structure or ternary strategy, the recent progress of tandem PSCs and ternary PSCs is summarized in Table 1. It is

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**Table 1**

The recent progress of tandem PSCs and ternary PSCs.

Tandem	Bottom Materials	PCE (%)	Top Materials	PCE (%)	Tandem PCE (%)	PCE* (%)	Ref.
	PTB7-Th:PC <sub>71</sub> BM	9.0	PTB7-Th:PC <sub>71</sub> BM	9.0	11.3	25.6	[24]
	PDTP-DFBT:PC <sub>71</sub> BM	7.6	PDTP-DFBT:PC <sub>71</sub> BM	8.1	10.2	25.9	[25]
	P3HT:IC <sub>60</sub> BA	5.7	PBDTT-DPP:PC <sub>71</sub> BM	6.5	8.6	32.3	[26]
	PIDT-phanQ:PC <sub>61</sub> BM	5.0	PIDT-phanQ:PC <sub>71</sub> BM	6.5	7.8	20.0	[27]
	PCDTBT:PC <sub>71</sub> BM	5.8	PDPP5T:PC <sub>61</sub> BM	5.3	7.0	20.7	[28]
	PCDTBT:PC <sub>71</sub> BM	6.25	PDPP3T:PC <sub>71</sub> BM	5.10	6.88	10.0	[18]
	PBDTTT-C-T:PC <sub>61</sub> BM	4.27	P3HT:IC <sub>60</sub> BA	5.24	6.18	17.9	[29]
Ternary	Binary Blends	PCE (%)	Complementary materials		Ternary PCE (%)	PCE* (%)	Ref.
	PBDTTPD-HT:PC <sub>71</sub> BM	6.85	BDT-3T CNCOO		8.40	22.6	[30]
	PTB7:PC <sub>71</sub> BM	7.25	PID2		8.22	13.4	[31]
	DTfBT:PC <sub>61</sub> BM	6.26	DTPyT		7.02	12.1	[32]
	P3HT:ICBA	4.68	QD:NCNT		6.11	30.6	[33]
	P3HT:PC <sub>61</sub> BM	3.27	DIB-SQ		4.51	37.9	[34]
	P3HT:PC <sub>71</sub> BM	3.90	TQTFa		4.50	15.4	[35]
	P3HT:PC <sub>71</sub> BM	3.35	SMPV1		4.06	21.2	[19]
	P3HT:PC <sub>61</sub> BM	3.10	THC8		3.88	25.2	[36]

PCE\* = PCE of tandem PSCs/the best PCE of sub-cells, or PCE of ternary PSCs/PCE of binary PSCs.

apparent that the percentage of PCE improvement by using tandem structure is similar to that of using ternary strategy. From device fabrication item, ternary strategy may provide a smart and effective method to improve the performance of PSCs based on a sandwiched structure. Up to now, most of the ternary PSCs are based on poly(3-hexylthiophene) (P3HT) as the major electron donor, which limits the optimized PCE of ternary PSCs. To further improve the PCE of ternary PSCs, the basic binary PSCs should exhibit a relatively high PCE based on narrow band gap material as electron donor.

In the past years, some highly efficient narrow band gap materials were successfully synthesized by using benzodithiophene (BDT), dithienosilole (DTS), or indacenodithiophene (IDT) donor units and benzothiadiazole (BT), thienopyrroledione (TPD), or thiazolothiazole (TTZ) acceptor units [37,38]. Guo et al. successfully synthesized a novel band gap polymer poly[[4,9-dihydro-4,4,9,9-tetra(4-hexylbenzyl)-s-indaceno[1,2-b:5,6-b']-dithiophene-2,7-diyl]-alt-[2,3-bis(3-(octyloxy)phenyl)-2,3-dihydro-quinoline-2,20-diyl]] (PIDTDTQx) and obtained a high PCE of 7.11% with [6]-phenyl-C71-butyric acid methyl ester (PC<sub>71</sub>BM) as the electron acceptor for the optimized PIDTDTQx:PC<sub>71</sub>BM doping weight ratio as 1:4 [39]. The small molecule material 2,4-bis[4-(N,N-diisobutylamino)-2,6-dihydroxyphenyl] squaraine (DIB-SQ) has been demonstrated as an efficient photovoltaic donor material due to its high charge carrier mobility and intense absorption in the longer wavelength range [40,41]. According to the absorption spectral range of polymer PIDTDTQx and small molecule DIB-SQ, both are the promising electron donor materials for obtaining high performance ternary PSCs. A series of PSCs were fabricated with PIDTDTQx as the main electron donor and DIB-SQ as the complementary electron donor, the only difference is the DIB-SQ doping ratio in donors. The champion PCE of ternary PSCs arrives to 6.49% along with a  $V_{OC}$  of 0.85 V, a  $J_{SC}$  of 11.56 mA/cm<sup>2</sup> and a FF of 66% when the DIB-SQ doping ratio is 9 wt% in donors. An 18.6% PCE improvement was obtained compared with the optimized binary PSCs based on PIDTDTQx:PC<sub>71</sub>BM as the active layers. The PCE improvement should be attributed to the better trade-off photon harvesting in the whole spectral range and the optimized exciton dissociation and charge carrier transport in the ternary active layers with appropriate DIB-SQ doping ratio in donors.

## 2. Experimental section

The indium tin oxide (ITO) substrates with a sheet resistance of 15 Ω/square were cleaned consecutively in the ultrasonic baths

containing acetone, detergent, de-ionized, and ethanol. The cleaned ITO-coated glass substrates were dried by high pure nitrogen gas and then treated by UV–ozone for 10 min to improve the work function of ITO glass substrates. Subsequently, poly-(3,4-ethylenedioxythiophene): poly-(styrenesulphonic acid) (PEDOT:PSS) (purchased from H.C. Starck co. Ltd.) solutions were spin-coated on the ITO-coated glass substrates at 5000 rounds per minute (RPM) for 40 s and baked at 120 °C for 15 min in air condition. Small molecule DIB-SQ and fullerene derivative PC<sub>71</sub>BM were purchased from Luminescence Technology Corp. and polymer PIDTDTQx was purchased from Organtec Materials Inc. The PIDTDTQx, DIB-SQ and PC<sub>71</sub>BM were separately dissolved in 1,2-dichlorobenzene (DCB) with the concentration of 10 mg/mL, 10 mg/mL and 40 mg/mL, respectively and solutions were heated and stirred in a high-purity nitrogen-filled glove box for 12 h. The blend solutions were prepared with the weight ratio of donor to acceptor as constant (1:4), the only difference is DIB-SQ doping ratio in donors from 0 wt% to 15 wt%. The prepared blend solutions with different DIB-SQ doping ratios were spin-coated on the PEDOT:PSS films at 2000 RPM for 40 s to fabricate the active layers in a high purity nitrogen-filled glove box. The prepared active layers were placed in covered Petri dishes for 5 min and then poly[(9,9-bis(3'-(N,N-dimethylamino) propyl)-2,7-fluorene)-alt-2,7-(9,9-dioctylfluorene)] (PFN) solution with a concentration of 0.2 mg/mL was spin coated on top of the active layers at 3000 RPM for 40 s. The combined cathode of Al (100 nm) was deposited with a shadow mask by thermal evaporation under 10<sup>-4</sup> Pa condition, and the thickness of each layer was monitored by a quartz crystal microbalance. The effective area of the device is 3.8 mm<sup>2</sup>, which is defined by the vertical overlap of ITO anode and Al cathode.

The absorption spectra of films were measured by a Shimadzu UV-3101PC spectrometer. The current–voltage (*I*-*V*) measurements of the devices were conducted using a Keithley 4200 source meter in air condition. AM 1.5G irradiation was provided by an ABET Sun 2000 solar simulator with light intensity of 100 mW/cm<sup>2</sup>. The external quantum efficiency (EQE) spectra of the PSCs were measured by a Zolix Solar Cell Scan 100. The thickness of the active layers was about 90 nm, measured by a Ambios Technology XP-2 stylus Profiler. Photoluminescence (PL) spectra of films were measured by a Perkin Elmer LS-55 spectrophotometer. Time-resolved transient photoluminescence (TRTPL) spectra were obtained using a FluoroCube-01-NL and FluoroCub-NL from Jobin Yvon. The morphology of films was investigated by atomic force microscopy (AFM) using a multimode Nanoscope IIIa operated in tapping mode. The chemical structure of used materials PIDTDTQx,

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