



Enhanced photovoltaic performance of ternary solar cells by doping a new squaraine derivative



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ABSTRACT

Ternary blend solar cells have been proved to be an efficient way to harvest more photons over the near-IR region. In this work, the effect of adding a NIR absorbing squaraine derivative (TPE-SQ) into a conventional P3HT:PC₇₁BM based bulk heterojunction photovoltaic cell is investigated. The new NIR dye exhibits a quite broad absorption band with full-width at half maximum of 169 nm in the film state, with the absorption edge extending to more than 850 nm. The near infrared absorption of the ternary system is enhanced by the blended squaraine component, and the energy level of the third component matched well with the donor and the acceptor material, leading to increased short-circuit current as well as elevated power conversion efficiency compared to the P3HT:PC₇₁BM binary solar cell. The highest efficiency of 3.93% is achieved for a P3HT:PC₇₁BM blend with 2.4 wt % of TPE-SQ.

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1. Introduction

As one of the most efficient technologies for solar energy conversion, polymer solar cells (PSCs) have attracted much attention because of their unique properties such as ease of processing, potentially low cost and large scale production. To date, the power conversion efficiency (PCE) beyond 10% has been achieved in PSCs with small active areas (~0.1 cm²), by altering polymer material, device architecture and improving fabricating techniques [1–3]. Typically, organic bulk heterojunction solar cells comprise a donor and an acceptor material blended together as the active layer, which is known as the binary system [4–6]. To further improve the PSC efficiency, one needs to enhance light absorption without sacrificing V_{oc} and FF , and more photons have to be harvested over a wide wavelength range from visible to near infrared (NIR) region. Whereas, it is generally difficult to cover such a wide absorption range by only two materials of conjugated polymer and fullerene, since the donor polymer mainly absorbs at the visible range while the fullerene hardly contributes to the absorption [7,8]. To address

this issue, tandem solar cells consisting of two or more single cells with complementary absorption ranges were designed, and impressively improved efficiency was gained [9,10]. However, the complex device fabrication of the tandem architecture impedes its development. In this regard, ternary blend solar cells incorporating two complementary donor materials into a single active layer have been proposed, which could extend the light-harvesting range from visible to the NIR region in a simple way.

Currently, two types of ternary blend solar cells are intensively studied in order to harvest more photons over the NIR region. One is based on two kinds of donor conjugated polymers blended with one acceptor material [11–17]. While the other one is composed of one donor polymer, one acceptor, and an additional NIR dye molecule [18–22]. The later approach has been demonstrated to be more convenient and versatile, which can be easily realized by using two different dye molecules with complementary absorption bands. In addition, conjugated small molecules tend to exhibit good miscibility with the electron donor and the electron acceptor materials, so as to control the domain size of the ternary BHJ active layer for efficient photoinduced charge transfer at the D-A interfaces. Therefore, in recent years, research on ternary BHJ-PSCs incorporating additional donors with NIR absorption has attracted much attention. Among the NIR dye molecules studied, the low-bandgap squaraine derivatives are mostly applied to the common binary blend of P3HT and PC₇₁BM system, and increased power

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conversion efficiencies as well as external quantum efficiencies have been obtained [23–26]. For example, Huang et al. used 2,4-bis[4-(*N,N*-diisobutylamino)-2,6-dihydroxyphenyl] squaraine (SQ) dye as the additional donor in P3HT:PC₇₁BM devices, and the PCE enhanced significantly from 3.27% in the reference cell to 4.51% in the device with 1 wt % SQ content [27]. Rao et al. studied the effect of incorporation of a squaraine dye TBU-SQ (bis[4-(2,6-di-*tert*-butyl)vinylpyrylium] squaraine) on the photovoltaic response of P3HT:PC₇₀BM system. It turned out that the improvement in PCE originated from the increased light-harvesting efficiency in the NIR region, and also the increased exciton dissociation into free charge carriers in the ternary blended film [25]. Later, An et al. reported similar results by adding SQ molecules into the P3HT:PC₇₁BM binary blend, leading to PCE increased from 3.05% to 3.72% with only 1.2 wt % SQ [23]. Meanwhile, new low-bandgap dye molecules have also been explored. As has been reported in the literature, low-band-gap small molecule DTCTB (2-((7-(5-(di-*p*-tolylamino)thiophen-2-yl)benzo[*c*] [1,2,5]thiadiazol-4-yl)methylene)malononitrile), phthalocyanine based heterostructured dye, and diketopyrrolopyrrole (DPP) derivatives were incorporated separately as the second donor material, and the PCE of the corresponding polymer solar cells were enhanced significantly with low doping contents of the NIR dyes [28–30]. Considering the advantages of the ternary systems, it is thus urgent and necessary to develop various NIR dyes and to investigate their application as the third component in ternary solar cells, aiming to further improve the power conversion efficiency of PSCs.

In this work, we demonstrate increased efficiency in a bulk heterojunction polymer solar cell device consisting of a blend of two donor materials, one is the most conventional polymer P3HT, and the other one is a new NIR small molecule based on a symmetric squaraine derivative ((*E*)-2-(1-hexyl-5-((*E*)-4-(1,2,2-triphenylvinyl)styryl)-1*H*-pyrrol-2-yl)-4-(1-hexyl-5-((*E*)-4-(1,2,2-triphenylvinyl)styryl)-2*H*-pyrrol-1-ium-2-ylidene)-3-oxocyclobut-1-enolate) (TPE-SQ) (Scheme 1). The devices are fabricated by doping the NIR donor TPE-SQ into P3HT mixed with the PC₇₁BM. We note that dye TPE-SQ exhibits strong absorption band from 600 to 800 nm with FWHM as large as 169 nm in the film state, while P3HT absorbs mainly from 400 to 630 nm. The highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular

orbital (LUMO) of TPE-SQ are positioned between the corresponding levels of P3HT and PC₇₁BM, respectively. Therefore, the cascaded energy level of P3HT:TPE-SQ:PC₇₁BM will enhance the exciton dissociation and charge carrier transport [31–33]. It turned out that by blending 2.4 wt % of the squaraine additive, both J_{sc} and PCE are increased compared to the device without TPE-SQ, and the optimized PCE of PSCs based on P3HT:TPE-SQ:PC₇₁BM is improved to 3.93%. Therefore, TPE-SQ could serve as a suitable third component to the P3HT:PC₇₁BM binary system.

2. Experimental section

2.1. Materials and instruments

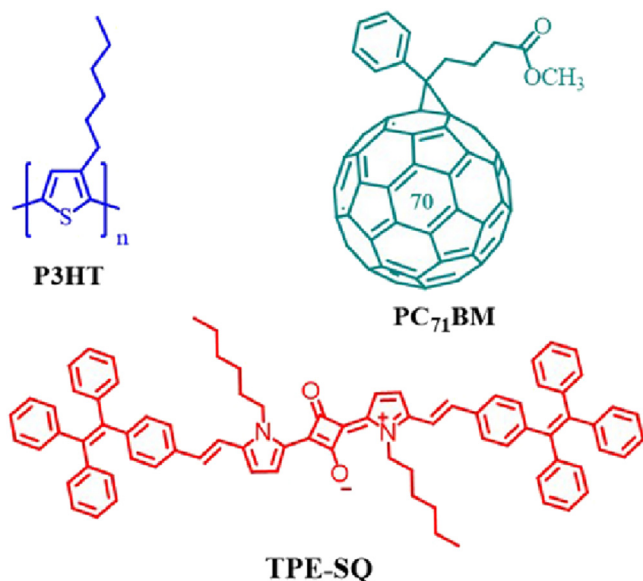
Unless otherwise stated, all starting materials were purchased from commercial suppliers (J&K Scientific and the Energy Chemical) and used without further purification. Tetrahydrofuran (THF) and toluene were freshly distilled over Na–K alloy under argon atmosphere prior to use. MALDI-TOF was performed on a Bruker Autoflex instrument, using 1,8,9-trihydroxyanthracene as a matrix. The NMR spectra were obtained from a BRUKER AVANCE III 600 MHz NMR Instrument (in CDCl₃). Elemental analyses of carbon, hydrogen, and nitrogen were performed on a Carlo Erba-1106 microanalyzer. The HRMS was measured on Thermo Scientific Q Exactive instrument. UV–vis absorption spectra were measured on a Shimadzu UV-2450 absorption spectrophotometer. Electrochemical experiments were performed using a CH Instruments electrochemical workstation (model 660A). The experiments were carried out in CH₂Cl₂ under argon atmosphere containing 0.1 mol/L tetrabutylammonium hexafluorophosphate (*n*-Bu₄NPF₆) as a supporting electrolyte at a scan rate of 50 mv/s. The potentials are quoted against the ferrocene internal standard.

2.2. Synthesis

General synthetic routes for the intermediate and the final compound TPE-SQ are depicted in Scheme 2. Diethyl 4-(1,2,2-triphenylvinyl)benzylphosphonate (**1**) and 1-hexyl-1*H*-pyrrole-2-carbaldehyde (**2**) were synthesized according to the methods reported in literature [34,35]. The final compound TPE-SQ has been fully characterized by ¹H NMR and ¹³C NMR spectroscopy, HRMS spectrometry, and elemental analysis as well.

2.2.1. Synthesis of compound 3

A solution of compound **1** (500 mg, 1.04 mmol) and compound **2** (186 mg, 1.04 mmol) in THF (40 mL) was cooled to 0 °C. NaH (150 mg, 3.75 mmol) was added in one portion under an Ar atmosphere. After stirring at 0 °C for 30 min, the mixture was warmed to room temperature and then was refluxed for 10 h. After cooling, the suspension was poured into ice water and extracted with CHCl₃. The organic layer was washed with brine and dried over anhydrous Na₂SO₄. After evaporation of solvents, the residue was purified by column chromatography over silica gel using petroleum ether/CHCl₃ mixture (5:1) as the eluent to give a bright yellow solid (320 mg, 61%). ¹H NMR (600 MHz, CDCl₃, ppm): δ 7.17 (d, *J* = 8.4 Hz, 2H), δ 7.15–7.01 (m, 15H), δ 6.97 (d, *J* = 8.4 Hz, 2H), δ 6.87 (d, *J* = 16.2 Hz, 1H), δ 6.76 (d, *J* = 15.6 Hz, 1H), δ 6.65 (t, *J* = 1.8 Hz, 1H), δ 6.44–6.43 (m, 1H), δ 6.13 (t, *J* = 3.0 Hz, 1H), δ 3.91 (t, *J* = 7.2 Hz, 2H), δ 1.73–1.70 (m, 2H), δ 1.33–1.26 (m, 6H), δ 0.87 (t, *J* = 6.6 Hz, 3H). ¹³C NMR (150 MHz, CDCl₃, ppm): δ 143.86, 143.80, 142.45, 140.92, 140.73, 136.04, 131.69, 131.44, 131.38, 127.79, 127.68, 127.63, 126.47, 126.40, 125.62, 125.24, 122.53, 116.88, 108.23, 106.56, 47.02, 31.55, 31.39, 30.90, 29.72, 26.48, 22.54, 13.99. MALDITOF-MS: *m/z* 507.3 (M⁺). Anal. calcd for C₃₈H₃₇N (%) : C, 89.90; H, 7.35; N, 2.76; Found: C, 90.27; H, 6.99; N, 3.06.



Scheme 1. Chemical structures of organic materials P3HT, PC₇₁BM and TPE-SQ.

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