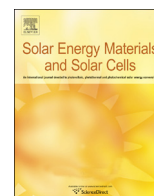




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All-polymer solar cells based on side-chain-isolated polythiophenes and poly(perylene diimide-alt-dithienothiophene)

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ABSTRACT

Two dimensional conjugated polymers have emerged as excellent polymer donor components in recent years. Here, all-polymer solar cells based on two conjugated side-chain isolated polythiophene derivatives (PT4TV and PT4TV-C) as donors and perylene diimide (PDI)-based polymer (PPDI-DTT) as acceptor are fabricated and investigated in detail. The structure similarity of the two polymers provides more insightful structure–property relationships. PT4TV based device displays a power conversion efficiency (PCE) of 0.99%, higher than that of PT4TV-C based device (0.55%). The high fill factor of larger than 50% in PT4TV based device is ascribed to the balanced hole and electron mobility of the donor and the acceptor. By adding chloronaphthalene solvent in PT4TV: PPDI-DTT solution, the PCE of PT4TV based solar cells is enhanced to 1.17%. Such improvement is interpreted in terms of the refined morphology of the film and ideality factor of the solar cell diode.

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1. Introduction

Polymer solar cells (PSCs) can be fabricated by spin coating, roll-to-roll and printing techniques on large-scale flexible substrates, this makes them very promising in photovoltaic applications [1–4]. With fullerene derivatives being used as acceptors, the power conversion efficiency (PCE) of bulk heterojunction polymer solar cells has reached 9.2% [5]. However, the capability of absorbing sun light for fullerene derivatives is generally very limited [6], which could eventually be detrimental to future PSCs based on them. Another consideration for developing n-type polymers for replacing commonly used fullerene acceptor is to overcome its open-circuit voltage (V_{OC}) limitation (ca. 1.1 V) [7]. In order to further improve the efficiency of PSCs, researchers start to employ n-type polymers with excellent absorption property and/or high LUMO (the lowest unoccupied molecule orbital) energy level as acceptors to replace the fullerene derivatives [8–13]. With its virtues of broad absorption (300–850 nm) and high electron mobility ($1.3 \times 10^{-2} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$), poly(perylene diimide-alt-dithienothiophene) (PDI-DTT) is a strong such acceptor candidate for using in all-PSCs [14].

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The structure diversity of the polymer donors and acceptors makes it easy to tune their energy levels and absorptions in PSCs, and hence to optimize the efficiency [15,16]. Despite of the exciting advantages of all-PSCs, the highest PCE of all-PSCs is only about 2% at present [10,17]. The low photocurrent (which seldom exceeds 5.0 mA cm^{-2}) and the low fill factor (which seldom exceeds 50%), are found to be responsible for the unsatisfactory performances. These two factors are related to the poor exciton dissociation at the donor/acceptor interface (which will strengthen bimolecular carrier recombination in PSCs [18]) and low carrier mobility [19] (which will hinder carrier transport and hence increase series resistance, a factor that determines fill factor of a solar cell).

The well-developed fullerene based PSCs can provide useful guidance for developing all-PSCs in terms of device engineering and the energy level matching between donor and acceptor. For the device engineering aspect, thermal annealing [20], additive [21,22] and mixture solvents [10,23] are proved to be efficient methods for optimization the blend morphology and thus the ultimate device efficiency. However, lessons from fullerene based device are not always suitable for all-PSCs. For example, some donor polymers work efficiently with fullerene acceptors, but their device performances with polymer acceptor are largely unpredictable despite of their matched energy levels. For example, linear conjugated polymer of poly(dithienosilole-benzothiadiazole) shows high efficiency of 5.1% with fullerene as acceptor.

While replacing fullerene with PDI-DTT, an undesirable low efficiency of 0.62% was obtained [22]. Notably, two dimensional conjugated polymers have emerged as excellent donor components in all-PSCs.

Two dimensional (2-D) conjugated polythiophene (PTs) derivatives (2-D-PTs) possess high hole mobility thanks to the 2-D conjugation structure, and broad absorption deriving from both the main chains and conjugated side chains, thus this family of 2-D-PTs demonstrated good device performances in PSCs. For example, using PDI-DTT as acceptors and (bis(thienylenevinylene)-substituted polythiophene as donor, prominent PCEs up to 1.5% were achieved. Subsequent simulations on these two polymer blends indicated that a PCE (power conversion efficiency) of 5% is reachable with an optimum of charge mobility and morphology. Later, Zhou et al. introduced longer conjugated side chains on the 2-D-PT backbone and developed new poly(perylene diimide)s as acceptors [10], recoded efficiency of 2.24% was achieved in 2011. Recently, Frechet and co-workers' investigations showed that the conjugated side-chain play an important role in all-PSCs [24]. By combination of computational and spectroscopic methods, their work suggested that the barrier to charge separation can be decreased by introducing conjugated substituent onto the polymer backbones, thus the photocurrent generation can be increased. This interesting result indicated that 2-D conjugated polymers have great potential in developing high efficiency all-PSCs. Thus investigation of new 2-D PTs with improved photo-physic properties can promise new opportunity to enhance the efficiency.

However, the above-mentioned 2-D-PTs showed poorer regioregularity thereby poor planarity and low hole mobility due to the asymmetric nature of 3-substituted thiophene units and high concentration of the substituents. To improve the planar of the backbone, we recently proposed a "side-chain-isolation" concept for the PTs and synthesized 2-D-PTs with unsubstituted terthienyl spacer, namely, PT4TV and PT4TV-C [25] (structure is shown in Fig. 1). The difference between the two polymers is an additional carbonyl group on the conjugated side chain in PT4TV-C. As a result, the planarity was largely improved. Compared to previously reported 2-D-PTs, the newly developed 2-D PTs showed red-shifted and enhanced π - π^* transition absorption of the polymer backbone, and higher hole mobility. Thus, with these new families of 2-D PTs as donors in all-PSCs, higher efficiency and more detailed structure-property relationship can be expected. Along this line, with PPDI-DTT as acceptor, we study the photovoltaic performance of these two new 2-D-PTs (PT4TV and PT4TV-C), various parameters of these all-PSCs are compared and studied in detail.

2. Experimental procedures

2.1. Materials and device fabrication

The device structure of these all-PSCs is ITO/PEDOT: PSS/polymer blend/Ca/Al. The fabrication processes are described in the following steps, the indium tin oxide (ITO) glass substrates were sequentially cleaned with deionized water, acetone, and isopropanol for 10 min each and then were treated by O_2 plasma for 6 min. A thin layer (ca. 35 nm) of PEDOT: PSS ((poly(3,4-ethylenedioxythiophene): poly-(styrenesulfonate)) was spin-coated on the pre-cleaned ITO-coated glass from a PEDOT: PSS aqueous solution (Baytron P VP Al 4083 from H.C. Starck) at 3000 rpm and annealed on a hotplate at 140 °C for 10 min in air, then the devices were transferred to N_2 glove box (< 0.1 ppm O_2 and H_2O), where the active layer of the mixed polymers was spin-coated. The optimized thickness of the active layers was ca. 55 nm. Finally the devices were transferred to a vacuum chamber

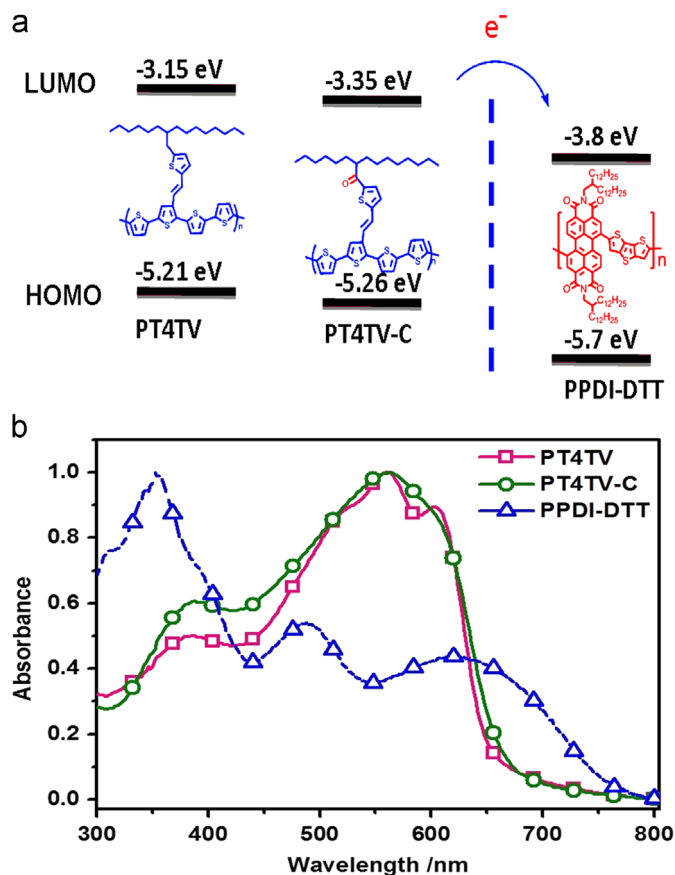


Fig. 1. (a) Chemical structures with the energy level diagrams of the two polymer donors (PT4TV and PT4TV-C) and the polymer acceptor (PPDI-DTT). (b) UV-vis absorption spectra of the three polymer thin films.

($< 10^{-5}$ Pa) to deposit a Ca (20 nm) layer and an Al (80 nm) layer with the active area of 0.17 cm^2 . The fabrication of active layers for transmission electron microscopy (TEM) measurement was similar to the photovoltaic devices. Then active layers with the ITO/PEDOT: PSS substrates were soaked in deionized water to separate the active layers from the substrates. The floated films were supported on the copper grids for the TEM measurement.

The OFET devices were fabricated in N_2 glove box. Gold electrodes were deposited on a 300 nm dielectric SiO_2 thermally grown on heavily doped n-type Si, and then PT4TV and PT4TV-C solutions (10 mg/mL) were spin-coated (2000 rpm, 30 s). OFET characters were measured by the equipment of Keithley 4200.

2.2. Characterization methods

The normalized UV-visible absorption spectra for all thin films spin-coated on quartz substrates were measured by a Hitachi U-3010 UV-vis spectrophotometer. Thickness of the active layer was determined by an Ambios Technology XP-2 surface profilometer. The current density-voltage (J - V) characteristics were measured under simulated sunlight of a 450 W Newport 6279 NS solar simulator. Simulated solar light was calibrated to match 100 $mW\ cm^{-2}$ AM 1.5G level by a standard mono-crystalline silicon solar cell. The incident-photon-to-converted-current efficiency was measured using Oriel Instrument's IQE-200.

The morphology of the active layers was carried out by atomic force microscopy (AFM) in air using the Multimode Nanoscope III equipment in tapping mode. A JEM 1011 TEM operated at 100 kV was employed for the bright field TEM images.

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