

Donor unit effect on DPP based organic field-effect transistor performance

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ABSTRACT

We report diketopyrrolopyrrole (DPP) based copolymers, poly(diketopyrrolopyrrole-cyclopentadithiophene) (PDPPTT-CPDT) and poly(diketopyrrolopyrrolebenzo[1,2-b:4,5-b']dithiophene) (PDPPTT-BDT) to investigate donor unit structure effects on organic field-effect transistor (OFET) performance. The highest occupied molecular orbital (HOMO) level and copolymer band gap were tuned by introducing different donor units on the acceptor DPP backbone. PDPPTT-BDT OFETs show 10 fold higher hole field-effect mobility, up to $0.08 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, than PDPPTT-CPDT OFETs due to better crystallinity and higher HOMO energetic levels. PDPPTT-CPDT and PDPPTT-BDT were operationally stable with negligible threshold voltage shift after cycling.

1. Introduction

Organic field-effect transistor (OFET) performance based on solution processed conjugated polymers has undergone significant progress recently as a result of combined efforts in developing novel materials [1], optimizing semiconductor morphology, improving charge injection property [2–5], developing favorable dielectrics [6–8], and controlling charge carrier density [9–11]. Low band gap donor-acceptor (D-A) copolymers, comprising alternating electron donating and accepting units, are promising classes of conjugated polymers for provision of high performance OFET polymer active layers [12–15]. Various functional groups have been investigated as donor or acceptor building blocks, and diketopyrrolopyrrole (DPP) is one of most widely studied acceptor units for D-A copolymers due to realizing high charge carrier mobility through large planarity and strong intermolecular π - π interaction [15–18].

When DPP and related electron accepting units associate with the appropriate electron donating unit, the resulting D-A molecules interact strongly between neighboring donor and acceptor units over short intermolecular π - π distances [19–22]. The large backbone planarity in DPP based polymers facilitates efficient intra-molecular charge transport through extended π -orbitals [23,24]. Utilization of intra- and inter-molecular charge transport has recently yielded impressive field-effect mobilities $> 10 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ in OFETs [25]. D-A molecules also exhibit efficient ambipolar charge injection from commonly used Au electrode due to their relative small bandgap via electron push and pull motif

[13,23]. However, partner donor unit structure effects on OFET performance should be further investigated to more accurately control DPP based D-A polymer charge injection and transport properties.

This study investigated the effect of cyclopentadithiophene (CPDT) and benzo dithiophene (BDT) donor units in DPP based D-A polymers on OFET performance using newly synthesized DPP based co-polymers poly(diketopyrrolopyrrole-cyclopentadithiophene) (PDPPTT-CPDT) and poly(diketopyrrolopyrrolebenzo[1,2-b:4,5-b']dithiophene) (PDPPTT-BDT) (see Fig. 1). CPDT and BDT are commonly used building blocks for conjugated molecules for organic solar cells [26,27] but have been rarely studied for OFETs. PDPPTT-BDT OFETs exhibited an order of magnitude higher hole mobility ($0.08 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) than PDPPTT-CPDT OFETs. X-ray diffraction (XRD) and ultraviolet photoelectron spectroscopy (UPS) suggest this large hole mobility difference is mainly due to high PDPPTT-BDT film crystallinity and higher HOMO energetic level providing better charge transport and injection.

2. Experimental

Reagents were purchased from Wako Pure Chemical Industries, Tokyo Kasei Chemical Industries, Merck, and Aldrich, and used without further purification. PDPPTT-CPDT and PDPPTT-BDT (compounds 4 and 5, respectively) were synthesized following procedures detailing in Supporting Information. Theoretical calculations for PDPPTT-CPDT and PDPPTT-BDT were conducted using the B3LYP functional and 6-31G (d, p) basis set as implemented in Gaussian09. Methyl group substituted A-D-A structures were modeled to shorten calculate time, and optimized

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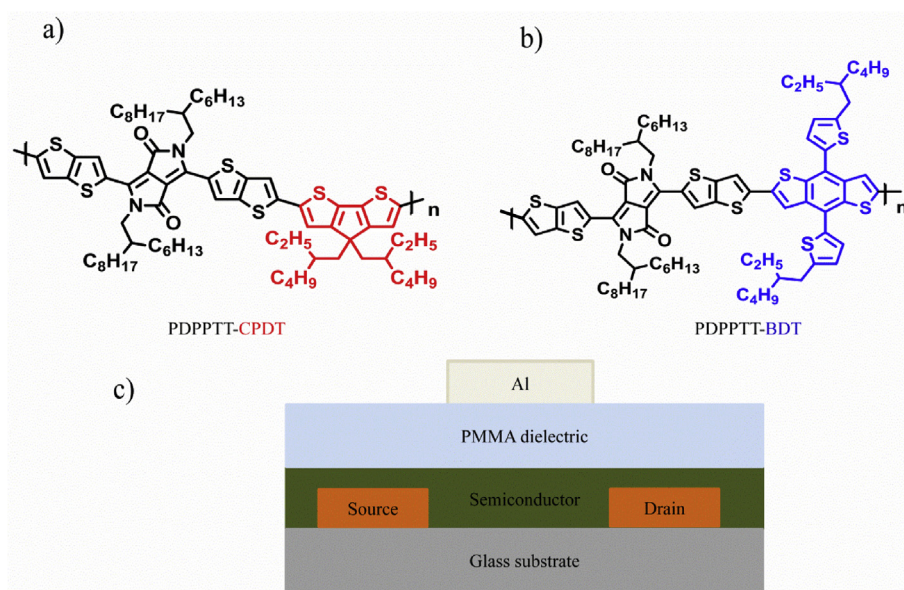


Fig. 1. Chemical structures for a) PDPPTT-CPDT and b) PDPPTT-BDT; and c) top gate/bottom contact organic field-effect transistors.

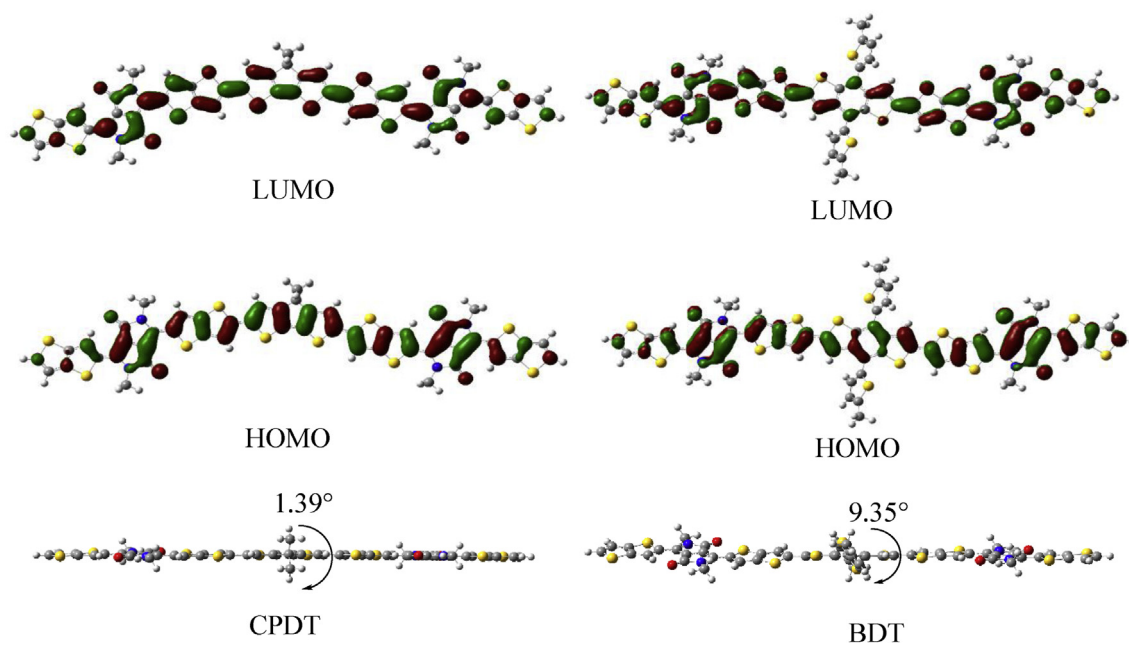


Fig. 2. Calculated molecular orbitals and side views with dihedral angles for PDPPTT-CPDT and PDPPTT-BDT (B3LYP/6-31G (d, p)).

structures are shown in Fig. 2. Polymer thermal stability was determined using thermogravimetric analysis under N₂ atmosphere.

Absorption spectra for diluted PDPPTT-CPDT and PDPPTT-BDT solutions (0.05 mg/mL) in 1,2-dichlorobenzene and thin-film samples were measured using a JASCO, V-770 UV-Vis-NIR spectrophotometer. To confirm polymer crystallinity, PDPPTT-CPDT and PDPPTT-BDT solutions in 1,2-dichlorobenzene were spin-coated on SiO₂ substrates and annealed at 150 °C, 200 °C, and 250 °C, for 30 min in an N₂ filled glove box. Thin film structural ordering and crystallinity were investigated using a Rigaku RINT 2000 X-ray diffractometer (XRD) with Cu K α radiation. ¹H NMR spectra were recorded with a JEOL ECA500 (500 MHz) spectrometer in chloroform-*d* (chemical shifts in parts per million (ppm) downfield from tetramethylsilane as an internal standard for ¹H). Column chromatography was performed with silica gel, (Kanto Chemical, silica-gel 60 N, 40–50 μ m).

Monomers for polymerizations were further purified by recycling

gel-permeation liquid chromatography (GPC) using a Japan Analytical Industry LC-908 equipped with JAI-GEL 1H/2H columns (eluent: CHCl₃). HPLC analysis was performed using a JASCO LC-NetII/ADC, JASCO PU-2080, JASCO UV-2077 and JASCO ChromNAV GPC systems with TOSOH TSKgel MultiporeH_{XL}-M column eluting with THF (1 mL/s) at 40 °C. Thermogravimetric analysis (TGA) was conducted by SII TG-DTA6200 connected with EXSTAR6000 under N₂ atmosphere.

Electrical performance was characterized by fabricating top gate/bottom contact (TGBC) staggered OFET devices. Source and drain electrodes (Au/Ni = 13/3 nm) were patterned on a glass substrate by photolithography and deposited onto the substrate through thermal evaporation. Electrodes were photo lithographically patterned to have 10–50 and 1000 μ m channel length and width, respectively. FeCl₃ (3 nm) was thermally evaporated as the hole injecting layer on the cleaned substrates. 5 mg/mL of PDPPTT-CPDT and PDPPTT-BDT solutions in 1,2-dichlorobenzene were spin coated at 1500 rpm for 150 s

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