

Organic thin-film transistors based on thieno[3,4-c]pyrrole-4,6-dione conjugated polymer via oxidative polymerization



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ARTICLE INFO

Article history:

Received 18 March 2013
Received in revised form 30 June 2013
Accepted 23 July 2013
Available online 3 September 2013

Keywords:

Thieno[3,4-c]pyrrole-4,6-dione
Oxidative polymerization
Organic thin-film transistors

ABSTRACT

Thieno[3,4-c]pyrrole-4,6-dione (TPD)-based polymer (PDTPD) with head-to-head (H-H) linkage of 3-dodecylthiophene unit was synthesized by ferric chloride oxidative polymerization. The polymer was characterized by chemical analysis as well as thermal analysis, optical spectroscopy and cyclic voltammeter. PDTPD polymer showed excellent thermal stability, had significantly red-shifted of absorption edge by its solid film compared to its solution and a deep highest occupied molecular orbital (HOMO) energy level of -5.82 eV. Polymer thin-film microstructures and morphologies were also investigated through atomic force microscope (AFM) and grazing-incidence X-ray diffraction (GIXD) which showed that the PDTPD film is composed of rod-like crystalline grains and ordered structures. Organic thin-film transistors (OTFT) with common architectures were fabricated to evaluate OTFT performance and exhibited the preliminary hole mobility of $0.019 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$.

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1. Introduction

Over the past decade, organic thin-film transistors (OTFT) based on conjugated polymers have undergone significant progress due to many advantages such as light weight, low-cost, and good compatibility with solution-processes that are promising for the fabrication of flexible large area devices [1–10]. To obtain higher charge mobilities, molecular ordering of the polymer semiconductors must be improved when processed from a solution [11–13]. Polythiophene derivatives such as poly(3-hexylthiophene) (P3HT), poly(2,5-bis(3-alkylthiophen-2-yl)thieno[3,2-b]thiophene) (PBTTT) possessed finely conjugated and coplanar conformation, and therefore possess a highly crystalline thin-film microstructure. The polythiophene-based device is reported to have charge mobilities as high as $0.1 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ with current on/off ratio above 10^6 [11–14]. However, these high-performance polythiophene derivatives were mostly constructed from electron-rich groups and possessed a relatively high energy level of the highest occupied molecular orbitals (HOMO), the air stability is still to be improved for practical applications. For

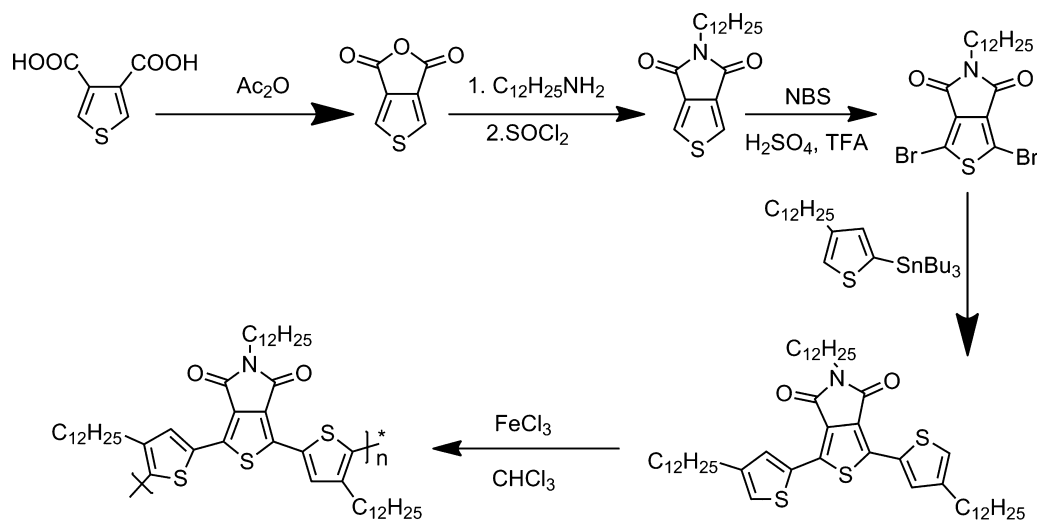
polymer semiconductors, lowering the HOMO energy levels will result in increasing the air stability. Therefore, fine tuning of the semiconductor energy levels may be the most effective way to improve their device stability [15–17]. So far, conjugated polymers with donor-acceptor (D-A) architectures have been an effective approach to enhanced the device stability. The D-A polymer which constructed by alternate electron rich and deficient units could more easily produced a low HOMO energy level and result in high air stability [15]. Recently, the D-A polymers based on thieno[3,4-c]pyrrole-4,6-dione (TPD) have received much attention due to its high performance in polymer solar cells and organic thin film transistors [18–26]. TPD possesses a compact, symmetric, and planar structure, the strong electron-withdrawing imide group of TPD greatly lowers the HOMO and the lowest unoccupied molecular orbital (LUMO) energy levels [21,25]. If the TPD unit is introduced into poly(3-alkylthiophene) backbone, the polymers would have lower HOMO energy levels and shows better air stability.

Taking all of these results into account, polymer (PDTPD) based on TPD and 3-dodecylthiophene units were firstly synthesized through ferric chloride oxidative polymerization in this paper (Scheme 1). The 3-dodecylthiophene units in PDTPD backbone form a head-to-head (H-H) conformation (Fig. 1). The thermal, optical, electrochemical properties and microstructure and morphology were also characterized in this work. The polymer PDTPD showed excellent thermal stability, and the film exhibited

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Scheme 1. Synthetic route of polymer PDTPD.

significant red shift of absorption edge compared to solution, which indicates a deep HOMO energy level of -5.82 eV. The polymer films on the substrates is composed of rod-like crystalline grains and ordered structure. The field-effect mobility was measured using OTFTs with a bottom-contact/bottom-gate configuration. Hole mobility of PDTPD-based devices as high as $0.019 \text{ cm}^{-2} \text{ V}^{-1} \text{ s}^{-1}$ is achieved.

2. Experimental

2.1. Instrumentation

Nuclear magnetic resonance (NMR) spectra were recorded on a Mercury plus 400 MHz machine. Gel permeation chromatography (GPC) analyses were performed on a Perkin Elmer Series 200 gel coupled with UV–vis detector using tetrahydrofuran as eluent and polystyrene as the standards. Thermogravimetric analyses (TGA) were conducted with a TA instrument Q5000IR at a heating rate of $20^\circ\text{C min}^{-1}$ under nitrogen gas flow. Differential scanning calorimetry (DSC) was performed on a TA instrument Q2000 in a nitrogen atmosphere. The samples (about 10.0 mg in weight) were firstly heated up to 388°C and held for 2 min to remove thermal history, followed by a cooling down at the rate of 20°C/min to 40°C and then a heating up at the rate of 20°C/min to 388°C . UV–vis absorption spectra were recorded with a Perkin Elmer model λ 20 UV–vis spectrophotometer. Electrochemical measurements were conducted under nitrogen in a deoxygenated anhydrous acetonitrile solution of tetra-*n*-butylammonium hexafluorophosphate

(0.1 M), using a CHI 660D electrochemical analyzer. The atomic force microscope (AFM, Multimode, Digital Instrument) operating in the tapping mode was used to characterize the surface morphology of films. The grazing-incidence X-ray diffraction (GIXD) studies were performed using 4C1 and 4C2 beamlines at Pohang University of Science and Technology.

2.2. Device fabrication and characterization of organic thin-film transistor (OTFT)

OTFT devices were fabricated with a bottom-contact/bottom-gate configuration. The device was fabricated on a heavily doped *n*-type Si wafer with a SiO_2 layer of 195 nm having a capacitance of 16.8 nF/cm^2 . The *n*-type Si layer served as the gate electrode and the SiO_2 layer acted as the gate dielectric. Octadecyltrichlorosilane (OTS) was used as a self-assembled surface modifier for the substrate. A solution of PDTPD in chloroform (6 mg/mL) was first filtered through a syringe filter, and then spin-coated on the surface-treated substrate at 1500 rpm for 20 s at room temperature. The semiconductor layer had a thickness of $\sim 150 \text{ nm}$ on the substrate. The OTFT devices had a channel width (W) of $250 \mu\text{m}$ and a channel length (L) of $10 \mu\text{m}$. The current–voltage (I – V) characteristics were measured with a Keithley 4200 SCS semiconductor characterization system. The carrier mobility in the saturated regime was calculated by using the equation $I_{DS} = (\mu WC_i/2L)(V_{GS} - V_T)^2$, where I_{DS} is the drain-source current, μ is the mobility, W and L are the semiconductor channel width and length, respectively. C_i is the capacitance per unit area

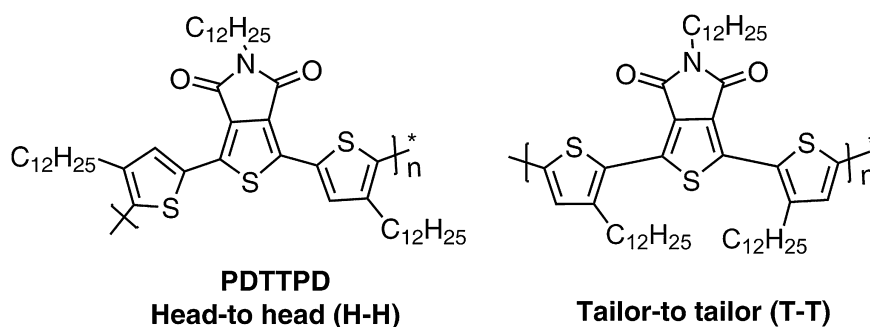


Fig. 1. Chemical structures of the H-H 3-dodecylthiophene containing polymer PDTPD and the T-T 3-dodecylthiophene containing polymer.

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