

Photo-response behavior of organic transistors based on thermally annealed semiconducting diketopyrrolopyrrole core

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ABSTRACT

We report the opto-electrical response of organic field-effect transistors based on a thin-film of a semiconducting diketopyrrolopyrrole (DPP) core, a popular building block for molecular semiconductors, and a polymeric gate dielectric. The thin-film of the DPP core was thermally annealed at different temperatures under N_2 atmosphere to investigate the relationship between the annealing temperature and the electrical properties of the device. The results showed that the annealing process induces morphological changes in the thin film, and properly controlling the thermal annealing conditions can enhance the device performance. In addition, we also investigated in detail the photo-response behaviors by analyzing the responsivity (R) of the device with the optimally annealed DPP-core thin film under two light illumination conditions by considering the irradiance absorbed by the thin film instead of the total irradiance of the light source. We found that the proposed model could lead to a light-source-independent description of the photo-response behavior of the device, and which can be used for other applications.

1. Introduction

Organic semiconducting materials containing extensive conjugated π -systems have been the focus of various research studies during the last few decades [1] for the development of organic based electronic devices such as organic field-effect transistors (OFETs) [2], organic photovoltaic (OPV) cells [3,4], organic light-emitting diodes (OLEDs) [5,6], and radio frequency identification (RFID) tags [7]. The main advantage of organic materials over inorganic ones is the potential to modify the chemical structure, which enables the appropriate selection of materials for specially designed applications such as flexible and stretchable electronics [8–10] or portable electronics with low power consumption [11].

Organic semiconducting materials usually have extensive π -orbitals in their backbone. Small molecules and polymers containing benzene, pyrrole or thiophene rings are widely used as active materials for various OFET or OPV devices [12,13]. Among them, diketopyrrolopyrrole (DPP) is a common π -conjugated moiety that appears in a wide range of polymeric [14,15], oligomeric [16] and small-molecular semiconductors [17,18]. The solubility of DPP-based compounds can be controlled by introducing alkyl chains to the N (position indicated by red diamonds in Fig. 1a) atoms of DPP core [19,20]. In general, DPP-based semiconductors are commonly substituted with a pair of thiophene rings that act as electron donors at the 3 and 6 positions, and

other aromatic groups can be introduced to the thiophene rings to modify the electrochemical properties (Fig. 1a). Modifying DPP derivatives to have extended π -conjugation and a more rigid structure would result in changes in the molecular packing, the highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) energy levels, and finally, the charge transport behavior of devices. Some examples include DPP derivatives containing 3-hexylthiophene [DPP(3HT)₂] [21,22], carbazole [DPP(CBZ)₂] [23] or cyano [DPP-T-DCV, DPP-CN] [24,25] groups. Although the molecular DPP derivatives with extended π -conjugation exhibit good charge transport behavior, it is quite important to use the simple soluble DPP core itself (non-functionalized soluble DPP, see Fig. 1b) as the semiconductor because synthesizing the abovementioned large DPP derivatives requires additional functionalization steps, resulting in a significant loss in the synthetic yield. Specifically, the synthetic yield can be lowered to around 60–70% [21,23,24], but in some cases it can be as low as 30% [26].

Although the non-functionalized soluble DPPs with an alkyl group have already been reported with conventional device performance parameters [19,20], other issues such as the effect of thermal annealing on device performance and the photo-response behavior must also be studied for various practical device application areas. In this paper, we report the effect of thermal annealing on the device performance of transistors based on a vacuum-evaporated DPP-R16 (2,5-dihexadecyl-

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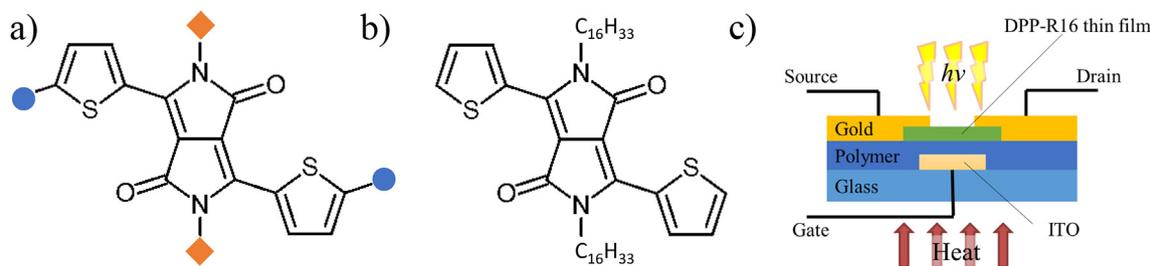


Fig. 1. a) The chemical structure of the diketopyrrolopyrrole (DPP) building block for the formation of its derivatives. A solubilizing group can be introduced at the positions indicated by red diamonds (◆), and additional functional moieties can be introduced at the positions indicated by the blue circles (●) in order to modify the electrochemical properties of the DPP core. b) The chemical structure of the non-functionalized soluble DPP core (DPP-R16) used in this study. c) Schematic structure of a transistor with the DPP-R16 thin-film active layer and a polymer gate dielectric. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

3,6-di(thiophen-2-yl)pyrrolo[3,4-c]pyrrole-1,4(2H,5H)-dione, Fig. 1b) thin film on a polymer gate dielectric (Fig. 1c), along with their operational stability under ambient conditions. More importantly, we report the photo-response behavior of the device, which is thermally annealed under optimum conditions, under different illumination conditions by considering the light source spectra, the absorption spectrum of the active layer, and the size of the active area to achieve a light-source independent description of the photo-response of the device. Thus, we propose a mathematical model and evaluate its applicability to devices measured under two different conventional polychromatic light sources.

2. Materials and methods

2.1. Synthesis and characterization of DPP-R16

The starting compound, to synthesize DPP-R16, i.e., 3,6-di(thiophen-2-yl)pyrrolo[3,4-c]pyrrole-1,4(2H,5H)-dione (pristine DPP), was prepared according to a procedure in the literature [27]. DPP-R16 was synthesized using the previously reported method [19]. A reaction mixture was prepared by dissolving pristine DPP, anhydrous K_2CO_3 and 18-crown-6 in anhydrous N,N -dimethylformamide, which was heated at 100 °C for 1 h under argon. A solution of n -bromohexadecane in DMF was added dropwise to the pristine DPP solution, and the reaction mixture was then further stirred at 120 °C for 18 h. After cooling to room temperature, 100 mL of water was added. The product was extracted from the aqueous layer by adding 100 mL of chloroform, and the extraction was repeated 3 times. The organic layer was then dried over $MgSO_4$ and filtered, and the solvent was removed. This crude product was purified using column chromatography using a 1:1 V/V chloroform-hexane mixture as an eluent. All the reagents and solvents for the synthesis were purchased from commercial suppliers, and purified according to standard procedures [28].

UV-visible spectrum of the DPP-R16 solution in 1,2-dichlorobenzene and of the solid state DPP-R16 film (50 nm thick on a glass substrate) was measured using a Varian Cary 100 spectrophotometer. X-ray diffraction (XRD) measurements were performed using a Rigaku Ultima IV X-ray diffractometer with a copper X-ray source ($\lambda = 0.154$ nm; operating at 40 kV, 40 mA). A fixed 10 mm wide divergence slit and a scintillation counter detector were used for the measurements. The tapping mode atomic force microscopy (AFM) images were recorded using Nanoscope IIIa (Digital Instruments) using RTESP-300 (Bruker) probe, taking 512 scan lines with 512 samples per line. Both XRD and AFM measurements were done with thin-films deposited over crosslinked poly(4-vinylphenol) (PVP) gate dielectric/glass substrate.

2.2. Device fabrication and characterization

Bottom-gate top-contact organic thin-film transistors (OTFTs) were

fabricated on glass substrates with a lithographically patterned indium tin oxide (ITO) gate electrode (Fig. 1c). The substrates were ultrasonically cleaned using a sequence of cleansing agents for 15 min each, i.e., a detergent solution, deionized water, acetone, and then dried in an oven. The ITO/glass substrates were treated with a UV-ozone cleaner prior to use. The gate dielectric precursor solution was prepared by mixing poly-(4-vinylphenol) (PVP), poly(melamine-co-formaldehyde) methylated (as a cross-linker, 84 w% in methanol), and propylene glycol monomethyl ether acetate (PGMEA), with a mass ratio of 10:5:85 (PVP:cross-linker:PGMEA). The precursor solution was spin-coated at 3000 rpm for 30 s onto the cleaned ITO/glass substrate, followed by soft baking on a hot plate at 90 °C for 15 min and hard baking at 175 °C for 60 min in a vacuum oven, resulting in a dielectric thickness of ca. 500 nm. The thickness of the film was confirmed by an XP-100 surface profilometer (Ambios Technology).

The 50 nm thick DPP-R16 thin-film was deposited onto the cross-linked PVP gate dielectric using thermal vacuum evaporation under a vacuum of 2×10^{-6} torr at a substrate temperature of 25 °C, at an evaporation rate of 3 Å/s. The thickness was monitored using a quartz crystal balance. Right after DPP-R16 deposition, the substrate was transferred to a glove box for thermal annealing at temperatures (T_{anneal}) of 50, 65, 75, 85 and 100 °C for 30 min. For simplicity, the thin films are labeled with a suffix to indicate the annealing temperature, e.g., DPP-R16-50 is the DPP-R16 thin film annealed at 50 °C. One substrate was separately prepared without a thermal treatment as a control. After annealing, the device was completed by depositing 50 nm thick gold source and drain electrodes through a shadow metal mask using thermal vacuum evaporation at the pressure of around 2×10^{-6} torr at 25 °C, at an evaporation rate of 3 Å/s. The channel length (L , 50 μm) and width (W , 1000 μm) of the device were determined by the dimensions of the metal mask. The device characteristics were studied using HP 4145 B and 4156 A Precision Semiconductor Parameter Analyzers. When measuring the transfer curves that serve as the basis for calculating the device parameters, the gate-to-source voltage was swept from $V_{GS} = -40$ V (ON state) to $V_{GS} = 20$ V (OFF state). The photo-response behavior of the DPP-R16 based phototransistor was studied on two separately fabricated devices: one was measured using a light-emitting diode (LED) lamp with a total irradiance of 12.54 mW/cm², the other one was measured using a halogen lamp with a total irradiance of 47.96 mW/cm². The spectrum and irradiance of light sources were measured by an Avantes AvaSpec-2048 \times 14 fiber optic spectrometer.

3. Results and discussion

The effect of thermally annealing the active layer on the device performance has been reported for polymer and small molecular semiconductors, and those studies revealed that the annealing process causes a structural rearrangement, resulting in changes in the crystallinity and charge carrier transport behaviors [29–34]. XRD

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