



# Narrow band-gap donor–acceptor copolymers based on diketopyrrolopyrrole and diphenylethene: Synthesis, characterization and application in field effect transistor



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## ABSTRACT

Three low band-gap diketopyrrolopyrrole based polymers with varying donor groups of furan, thiophene and phenyl were synthesized and then copolymerized with diphenylethene. We investigate the influence of different donor groups and comonomers on the band-gap and field effect transistors. The efficient synthesis of the diketopyrrolopyrrole based copolymers was clearly characterized by a variety of measurements. Two dimensional Grazing Incident X-ray Diffraction was measured to prove that furan and thiophene based copolymers have ordered edge-on structure. These copolymers exhibited strong  $\pi$ – $\pi$  stacking and excellent hole mobilities when applied in the electric double layer field effect transistors. The high mobility of  $2.36 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  with an on/off ratio of  $10^3$  was achieved.

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## 1. Introduction

In recent years, donor–acceptor (D–A)  $\pi$ -conjugated polymers with narrow-band-gap have emerged as promising candidates for optoelectronic devices [1–8]. Diketopyrrolopyrrole (DPP) was mostly used as an electron acceptor to form D–A copolymers for its highly conjugated lactam structure, electron withdrawing capability and a high degree of planarity [9,10]. The polymers involving DPP units have showed enhancing intramolecular charge transport via extended delocalized  $\pi$ -electron distribution along the backbone [11–15]. The low band-gap which originates from coplanarity and well conjugated structure of DPP based copolymers makes these polymers as the key materials for semiconducting application [16–20].

Molecular packing of donor–acceptor has a great effect on the performance of electronic device [21–25]. By carefully selecting the electron donating groups on the lactam ring or the comonomer, its highest occupied molecular orbitals (HOMO) and lowest occupied molecular orbitals (LUMO) can be tuned which leads to innovative low band-gap materials [26,27]. Various electron donating groups,

such as thiophene, phenyl, selenophene and furan have been employed in DPP based copolymers [28–30]. DPP based copolymers can be prepared with the desired comonomer via either Suzuki coupling or Stille coupling [31–35]. Variation of the comonomer provides vast possibilities with different unit combination which may cause promising properties for semiconducting devices [36–38]. The enthusiasm towards various DPP based polymers accelerates the development of many applications in the polymer solar cells (PSCs) and field effect transistors (FETs) [39–43]. For instance, Sonar and co-workers have reported three DPP based copolymers using furan as the donor group and alternatively using phenylene, naphthalene or anthracene as donor comonomer blocks which finally achieved a mobility of  $0.11 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  [44]. Shahid and co-workers have reported the synthesis of two alternating copolymers of thiophene and selenophene flanked DPP with dithienothiophene which exhibited great charge transport properties with high mobility up to  $0.23 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  [29]. Yun and co-workers have reported that by introducing nitrile group as an electron withdrawing unit and vinyl as linkage in the DPP-based copolymer, a high mobility of  $7.0 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  was obtained when applied in field-effect transistors [45].

We are interested in systematically investigating the effects of different flanking block and comonomers on lowering the band

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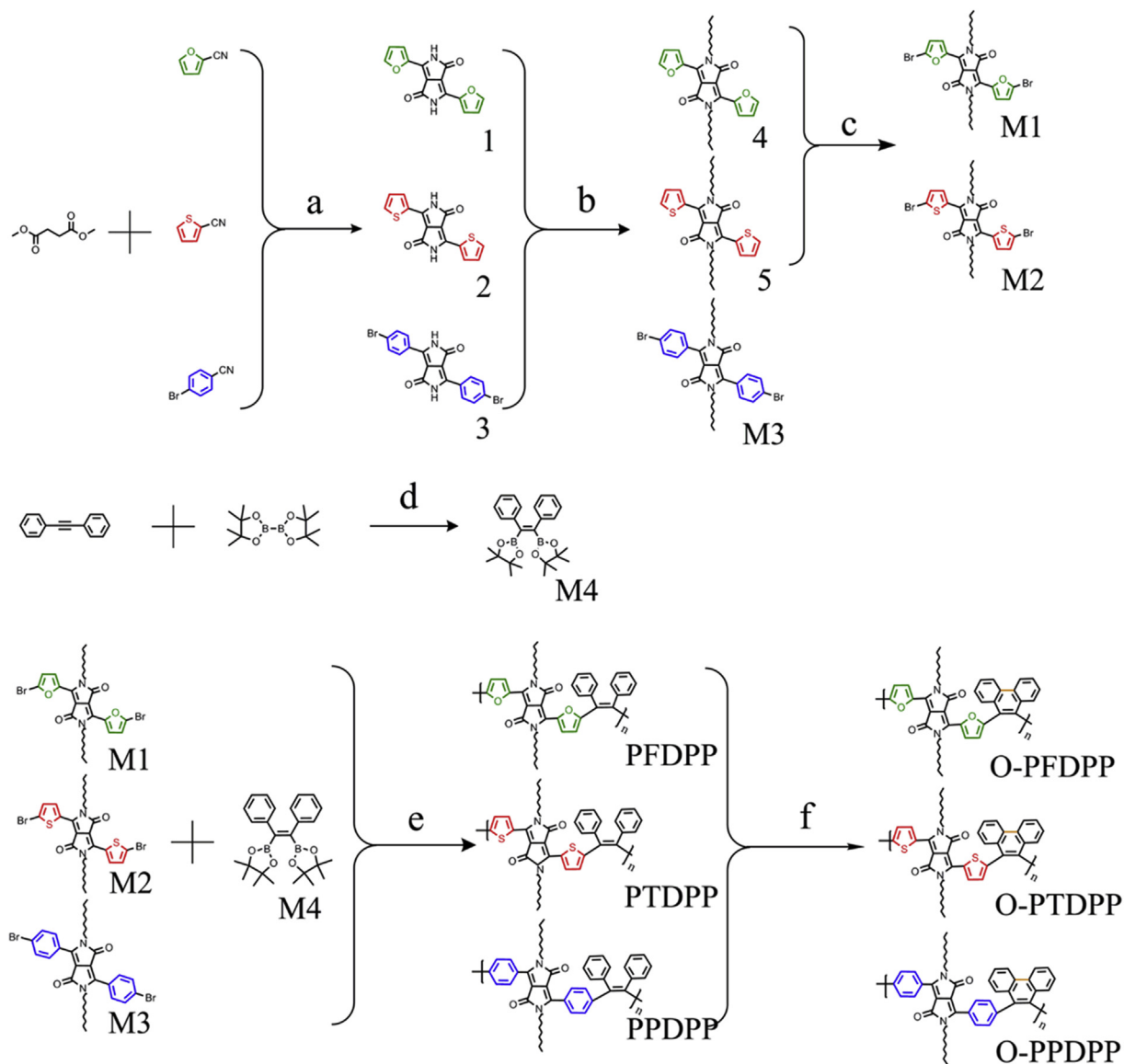
gap. Here, furan, thiophene and phenyl were used to attach on the DPP electron accepting moiety and then copolymerized with diphenylethene units which were shorted as **PFDPP**, **PTDPP** and **PPDPP**. Diphenylethene have been used in many materials with high charge carrier mobilities [46,47]. The chemical structures of three DPP based copolymers **PFDPP**, **PTDPP** and **PPDPP** are shown in Scheme 1. The values of aromatic resonance energy for furan, thiophene and phenyl are 0.70 eV, 1.26 eV and 1.56 eV, respectively. The electron donating strength suggests the effective conjugation length and enhancement of planarity which would decrease the band-gap of organic semiconducting devices [48]. With diphenylethene units, DDP derivatives provide a longer effective conjugation length and promote a more delocalized HOMO distribution along the backbone which would enhance the intermolecular charge-carrier hopping [35]. To further study the electron donating comonomers, diphenylethene was oxidized to become a rigid structure. The three oxidized copolymers **O-PFDPP**, **O-PTDPP** and **O-PPDPP** are shown in Scheme 1. Lei and co-workers have substituted the donor part of conjugated polymers which showed

the rotatable donor groups provided better FET performance than the rigid donor groups [24]. Here we demonstrate that comonomer with rigid structure leads to a larger band gap due to worse  $\pi$ - $\pi$  stacking and intermolecular interaction which originates from the increasing torsional angle. The copolymers were characterized by nuclear magnetic resonance spectroscopy (NMR), gel permeation chromatography (GPC), thermogravimetric analysis (TGA), UV-vis absorption and cyclic voltammetry (CV). **PFDPP**, **PTDPP** and **PPDPP** based electric double layer transistors exhibited a p-channel performance with high mobilities. The peak mobility value of  $2.36 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$  can be reached when **PFDPP** was applied to field effect transistors.

## 2. Experimental

### 2.1. Materials and instruments

Unless otherwise stated, all of reagents and chemicals used were purchased from Aldrich or Sinopham Chemical Reagent Company.



**Scheme 1.** Procedures for the synthesis of the monomer and the polymer. Conditions: (a) *t*-BuOK, *t*-amyl alcohol, 110 °C, 4 h, under argon; (b) 2-ethylhexyl bromide, DMF, potassium carbonate, 145 °C, 15 h, under argon; (c) NBS, chloroform, rt, 40 h; under argon; (d) Pt(PPh<sub>3</sub>)<sub>4</sub>, DMF, 90 °C, 24 h; (e) Pd(PPh<sub>3</sub>)<sub>4</sub>, 2 M K<sub>2</sub>CO<sub>3</sub>(aq), toluene, 110 °C, 8 h; (f) FeCl<sub>3</sub>, CHCl<sub>3</sub>, 3 min.

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