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Article

Theoretical and Experimental Study of Different Side Chains on 3,4-Ethylenedioxythiophene and Diketopyrrolopyrrole Derived Polymers: Towards Organic Transistors

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Abstract: In this research, two polymers of P1 and P2 based on monomers consisting of thiophene, 3,4-Ethylenedioxythiophene (EDOT) and diketopyrrolopyrrole (DPP) were designed and obtained by Stille coupling polycondensation. The material shows excellent coplanarity and structural regularity due to the fine planarity of DPP itself and the weak non-covalent bonding interactions existing between the three units. Two different lengths of non-conjugated side chains are introduced, which have an effect on the intermolecular chain stacking causing the film absorption to display different characteristic properties. On the other hand, the difference in side chains does not have a significant effect on the thermal stability and the energy levels of the frontier orbitals of the materials, which is related to the fact that the materials both feature extremely high conjugation lengths and specific molecular compositions. Microscopic investigations targeting the side chains provide a contribution to the further design of organic semiconductor materials that meet device requirements. Tests based on organic transistors show a slight difference in conductivity between the two polymers, with P2 having better hole mobility than P1. This study highlights the importance of the impact of side chains on device performance, especially in the field of organic electronics.

Keywords: copolymers; Stille coupling polycondensation; coplanarity; conjugated and non-conjugated; hole mobility

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1. Introduction

In recent years there have been numerous reports on the research of organic materials as new semiconductor materials in the field of organic electronics, which is directly related to the chemically modifiability of organic materials [1–4]. Functionalized device applications in the field of organic electronics generally include organic light-emitting, organic solar cells, organic electrochemical transistors, organic thermoelectrics, organic sensors, organic field-effect transistors (OFETs) and so on [5–8]. In order to realize functionalized applications and continuously improve device performance, it is necessary to design and prepare novel materials with suitable molecular structures [9–13]. A variety of organic molecular structures with the advantages of light weight, flexibility, low cost, and solution processability have been developed, especially for organic polymer materials [14–16]. One class of widely studied systems is based on polythiophene, which has good electron-donating ability and serves as one of the most essential materials for hole transport. In the case of poly(3-hexylthiophene) (P3HT), for example, the polymerization process is affected by the catalyst within and other experimental conditions. It is often

criticized for its tendency to form a variety of isomers such as head-tail and head-head, which affects the regularity of the material [17,18]. The coplanarity of the molecular structure in the material facilitates the formation of good stacking and promotes intra- and inter-chain transport of carriers to improve the performance of the device [19,20]. To improve the overall planarity of the backbone, various methods based on chemical molecular structure modification have been developed. First, conformational control groups such as alkenes and alkynes can be inserted between thiophene and thiophene to limit the torsion of thiophene. A typical example is the thiophene-vinylene-thiophene (TVT) structure [21,22]. Liu et al. reported a polymer of the DPP-TVT class with very close $\pi - \pi$ stacking distances and good electron mobility [23]. Thiophene can then also be fused with other aromatic rings to form heteroaromatic groups, such as benzothiophene and selenothiophene, in order to improve the coplanarity of the backbone [24]. Biewer et al. recently reported polymers based on the composition of DPP and thieno [3,2-b]pyrrole and showed $10^{-2} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ of moderate hole mobility. Finally, the formation of intramolecular conformational locks through weak intramolecular forces is also a method of interest. Intramolecular hydrogen bonding, S...O forces, S...F forces, electrostatic potential gravity, van der Waals forces, etc. are potential building systems [25–27]. Weak interaction forces tend to be flexible compared to TVT structures or heteroaromatic structures with rigid architecture. Weak interaction forces and weak crystallinity are often required for self-recovering and self-repairing functions in materials and devices oriented towards stretchability or foldability, thus achieving a balance between electrical and mechanical properties [28]. Wang et al. prepare stretchable transistor devices by physically mixing semiconductor conjugated polymer P3HT-like components with elastomeric components [29]. Hsu et al. provide ideas for soft electronic devices through a soft-end-hard-end combination strategy [30].

In this study, we have further expanded the electron-donating electrons of thiophene monomers by introducing electron-rich group of 3,4-Ethylenedioxythiophene (EDOT) [31–34]. At the same time, we expect to use the introduced O atoms to form more intramolecular weak forces to control the conformation and planarity of the materials. We used the structure of EDOT with a common acceptor building block diketopyrrolopyrrole (DPP) to prepare the polymers P1 and P2 by the Stille polymerization [35–37]. Studies in recent years have also demonstrated the direct effect of side chains on material properties and device performance [38,39]. The objective of this study was to investigate the effect of side chains on the physical properties and electrical conductivity of such polymers, based on the use of two DPP units with different alkyl chains (R).

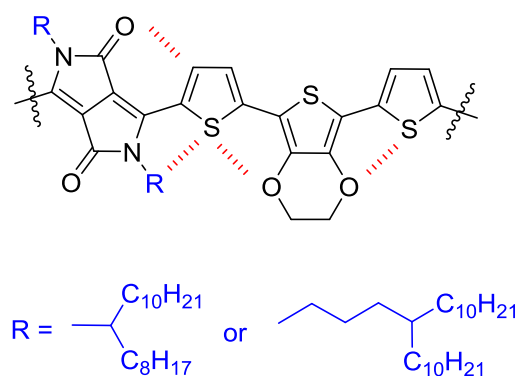
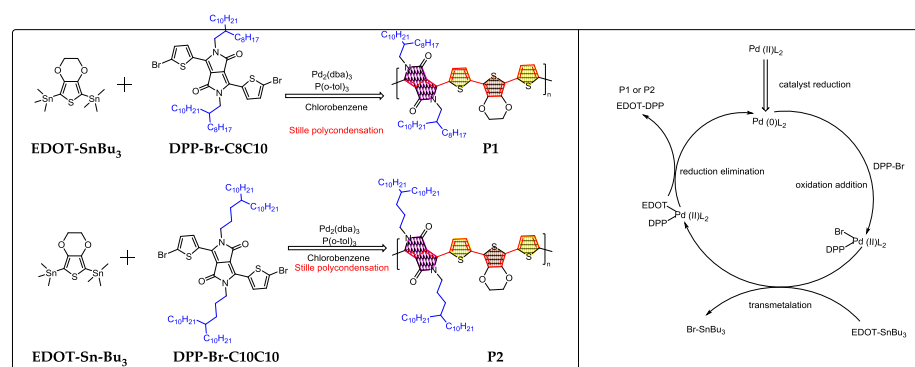


Figure 1. Stabilization of molecular configurations by introducing weak intramolecular forces as a strategy to prepare new structural motifs.

2. Results

2.1. Synthesis Routes to Polymers P1 and P2 through Stille Coupling Polymerization

The synthetic route for the preparation of the polymer by a palladium (Pd) catalyzed Stille coupling reaction of the two monomers is shown on the left Scheme 1. The tris (dibenzylideneacetone) dipalladium is used as the catalyst, often abbreviated to $\text{Pd}_2(\text{dba})_3$. In order to stimulate the catalyst's activities, the addition of a phosphorus ligand is required to reduce the divalent palladium to a zero-valent one. Subsequently, the C–Br bond in the monomeric DPP-Br-C8C10 or DPP-Br-C10C10 breaks readily and undergoes an oxidative addition reaction with the Pd coordinates, which initiates the reaction and starts the cycle (right side of Scheme 1). The choice of chlorobenzene solvent was related to the solubility of the polymer. While the DPP monomer showed good solubility in conventional solvents due to its long alkyl side chains, the polymer was only soluble in chlorinated solvents due to its extremely high molecular weight. The same choice of chemical equivalents and reaction time controls were used to analyze the differences in the final products of the reaction. The polymerization reaction was terminated after 12 h and purified by Soxhlet extraction [40]. The low molecular weight oligomers were extracted sequentially through methanol, acetone and hexane. The target materials were finally collected in chloroform in 88% and 79% yields, respectively. Polymers P1 and P2 showed high solubility in halogenated solvents such as chloroform or chlorobenzene with a dark green color and limited solubility in common organic solvents such as ethyl acetate. The solubility in both chloroform and chlorobenzene exceeded 5 mg/mL, so these solvents were used for subsequent processing. The molecular weights of the materials were characterized using high temperature gel permeation chromatography and the results are summarized in Table 1. The dispersive indexes (\mathcal{D}) of the two polymers were comparable, but their number average molecular weights (M_n) and weight average molecular weights (M_w) varied considerably. The viscosity average molecular weight (M_v), the Z average molecular weight (M_z), the peak average molecular weight (M_p) and the Z + 1 average molecular weight (M_{z+1}) of the polymers are listed in Figure S1 in Supplementary Materials and are shown in the usual order $M_n < M_v < M_w < M_z < M_{z+1}$. The molecular weight of polymer P1 was found to be approximately twice that of P2 (M_w of 54.4 k and 27.4 k, respectively). Based on the ratio of M_w to the molecular weight of the smallest repeating unit in the polymer chain, the average chain lengths of the two polymers are assumed to be 54 and 24, respectively. Elemental analysis of the polymer solids verified the composition and purity of the materials, the results of which are shown in Table 1 below.



Scheme 1. (left) Synthesis of polymers P1 and P2 based on the palladium-catalyzed polycondensation reaction, where the single and double bond conjugated fractions are shown alternately in red; (right) Sketch of the cycling mechanism based on a palladium catalyst.

Table 1. Molecular weight of P1, P2 and the ratio of three elements it contains.

M_n	M_w	\mathcal{D}^1	C	H	N
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				(%)	(%)	(%)
P1	15,419	54,417	3.53	72.94	9.98	1.22
P2	8204	27,456	3.35	73.57	8.77	2.72
repeating unit ²	1001	1001	N/A	71.95	9.26	2.80
repeating unit ³	1113	1113	N/A	73.33	9.77	2.52

¹ $\bar{D} = M_w/M_n$; ² The molecular chemical formula is $C_{60}H_{92}N_2O_4S_3$; ³ The molecular chemical formula is $C_{68}H_{108}N_2O_4S_3$.

In order to compare the thermodynamic stability of the two polymers, thermogravimetric analyses (TGA) and differential scanning calorimeter (DSC) were carried out and the curves are shown in Figures 2 and S2, respectively. Although the polymers have different chain lengths, they both exhibit similar stability in the range of 30–370 °C with very little decomposition. Specifically, P1 loses 5% of its weight at temperatures approaching 380 °C, while P2 loses 5% of its weight at 385 °C. We believe that this small difference may be related to the length of the side chains of the material. The thermal stability of conjugated materials beyond a certain length of conjugation is mainly related to the molecular composition of the side chains, but is not proportional to the length of conjugation [41]. The results of the infrared (IR) patterns are shown in Figure 2c,d below, where the IR absorptions of the materials with different conjugation lengths are similar. Their typical carbon-oxygen (C–O) bonding and carbonyl characteristic (C=O) peaks appear near 1080 cm^{-1} and 1655 cm^{-1} , respectively. Characteristic peaks of long aliphatic chains are obvious and found in the range of 2850–2920 cm^{-1} .

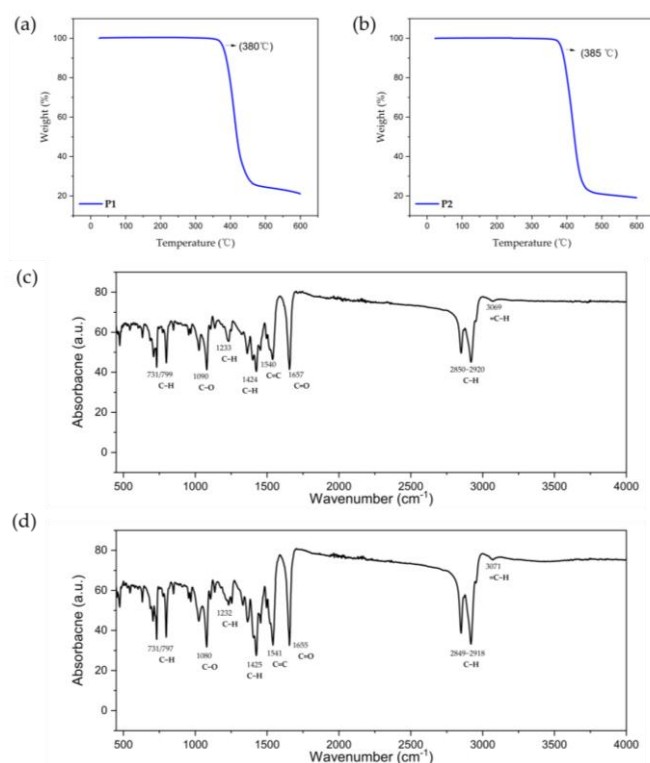


Figure 2. TGA curves of polymers (a) P1 and (b) P2; IR spectrum of polymers (c) P1 and (d) P2.

2.2. Photochemical Properties

The ultraviolet-visible absorption of P1 and P2 was carried out in dilute solution and in the solid state of the thin film, respectively, and the results are shown in Figure 3 below. Their absorption phenomena in solution are almost the same and both exhibit two absorption bands, which is due to the same molecular composition of the conjugated structure of the two materials. The relatively low absorption band in the 350–500 nm

range originates from the π \rightarrow π^* transition, while the significantly higher absorption intensity in the 700–1100 nm range is due to the intramolecular charge transfer from the donor to the acceptor. P1 displays a maximum absorption (λ_{\max}) peak at 952 nm and a shoulder peak near 874 nm in solution. The λ_{\max} peak of P2 in solution does not deviate from the maximum absorption peak of P1 by more than 5 nm (Table 2). On the other hand, the absorption of the two materials in the film phase is quite different. P2 still exhibits a maximum absorption peak as well as a pronounced shoulder peak, whereas P1 does not feature the tiny shoulder. The onset of absorption (λ_{onset}) in the solid state produces a redshift of approximately 50 nm with respect to the λ_{onset} in the solution state, which leads to a significant decrease in the energy level. The energy bandgap of P2 in the solid-state film is only 1.12 eV, which indicates a better π - π stacking in the film state.

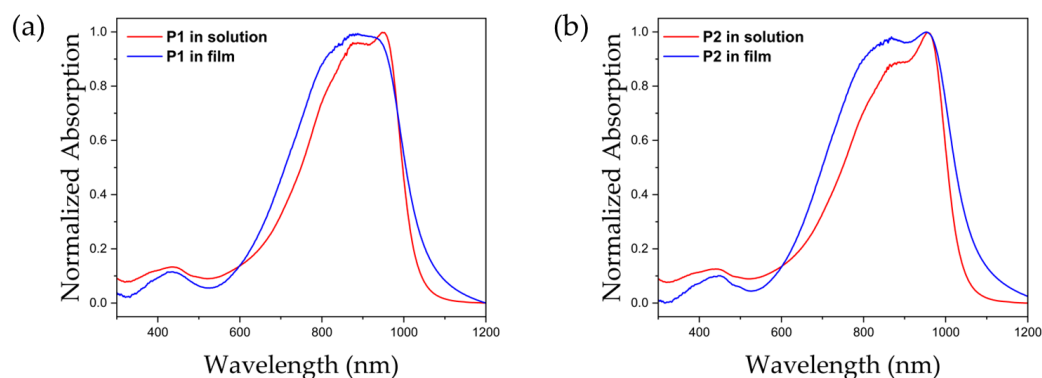


Figure 3. The ultraviolet-visible absorption of (a) P1 and (b) P2 in chloroform solution (0.5 mg/mL) and in thin film.

Table 2. Absorption peak data for polymers in solution and film phases.

Materials	λ_{\max}	λ_{onset}	E_g^1
P1 in solution	874, 952	1052	1.18
P2 in solution	873, 955	1049	1.18
P1 in film	887	1095	1.13
P2 in film	870, 955	1103	1.12

¹ $E_g = 1240/\lambda_{\text{onset}}$.

2.3. Electrochemical Properties

In order to further investigate the effect of varying side chains of the materials on their redox properties and the energy levels of the frontier molecular orbitals (FMO), electrochemical tests based on cyclic voltammetry were conducted as shown in Figure 4 below. Both polymeric materials show quasi-reversible oxidation characteristics with peaks of 0.99 V and 1.01 V. Simultaneously, they exhibit similar reduction profiles and possess peaks of -0.93 V and -0.94 , respectively. Depending on the onset of the oxidation peak and the onset of the reduction peak (0.77 V and -0.66 V, respectively), we can estimate that the corresponding highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) energy levels of P1 are -5.17 eV and -3.74 eV, respectively. The energy levels of the HOMO and LUMO of P2 are extremely close to those of P1 at -5.20 eV and -3.76 eV, respectively. Our results from this test show that despite the slight differences in the side chains, they do not have a drastic effect on the FMO energy levels of the materials. Aliphatic side chains consisting of non-conjugated components do not contribute significantly to the energy level of the conjugated main chain fraction [39,42].

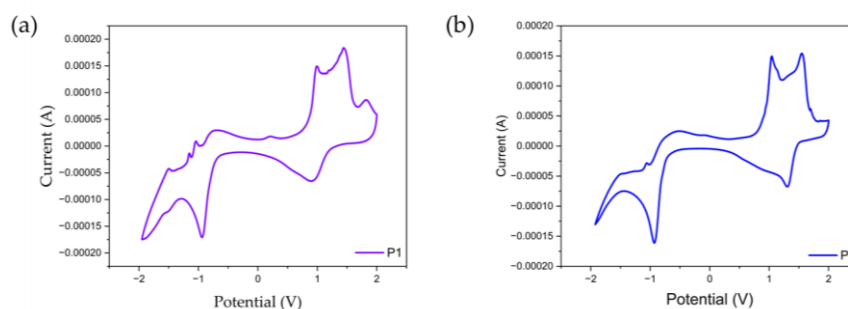


Figure 4. Redox characteristic of π -conjugated polymer P1 and P2 in the thin film (scanning rate: 0.1 V/s).

2.4. Density Functional Theory (DFT) Calculations

To further characterize the intramolecular conformation of the polymers, DFT calculations were used based on Gaussian 16 software [43]. For geometry optimization and frequency calculations, the B3LYP functional and the def2-SVP basis set was used, and the optimal geometry for each compound was determined [44,45]. The additional DFT-D3 London dispersion was introduced and analyzed in order to obtain accurate details of the intramolecular structure [46]. We used isopropyl chain-substituted dimers as computational models in order to balance the computational cost with the need to maximize the microscopic contribution of side chains to the structure [47]. Representative intramolecular atomic distances and dihedral angles between fragments presented in Figure 5a and 5b, respectively. The DPP unit consists of two five-membered fused aromatic rings and exhibits good coplanarity. The DPP ends are linked to the thiophene by a single bond and exhibit a small dihedral angle of about 16° . The distance between the oxygen atom (O) in the carbonyl group and the hydrogen atom (H) in the neighboring thiophene is only 2.1 Å, which is smaller than the van der Waals force radius and thus facilitates the formation of intramolecular hydrogen bonds. The sulfur atom (S) on thiophene shares a S...O distance of 2.9 Å with EDOT, which facilitates the formation of an intramolecular conformational lock and thus further stabilizes the conformation of the material. The weak non-covalent bond interactions analysis in Figure 5c further validates the attraction of the stable configuration present within the molecule. Intramolecular reduced density gradient (RDG) analyses revealed a pronounced bluish-green coloration across their corresponding positions, demonstrating these attractive interactions [48]. The oxygen of the carbonyl group forms an intramolecular hydrogen bond with the hydrogen atom on thiophene, which appears blue-green in color. The S atoms have a pronounced van der Waals gravitational attraction with the H atoms on the alkyl chain. The thiophene unit interacts with the EDOT unit via the S...O interactions and S...H interactions to form a flat coplanar structure. The electrostatic surface potential (ESP) of the material is characterized in Figure 5d, where the carbonyl position is surrounded by a clear attraction of electrons whereas the EDOT ring shows a donation feature. The results of the orbital energy level analysis of the dimer are depicted in Figure 5e below, and its corresponding HOMO and LUMO energy levels are calculated to be -5.10 eV and -3.51 eV, respectively. It is worth mentioning that the HOMO energy levels calculated from theoretical simulations are close to those derived from electrochemical tests. The theoretical and experimental values of the LUMO energy levels are slightly different, resulting in a difference of about 0.15 V between the bandgaps obtained by the two techniques. The orbital diagrams and of the corresponding HOMO - 1/LUMO + 1 and HOMO - 2/LUMO + 2 are placed in Figure S3. Figure S4 demonstrates the theoretically simulated absorption characteristic curve of the dimer, accompanied by the maximum absorption peak appearing near 815 nm. The onset absorption peaks are in close agreement with the actual absorption measurements, both appearing in the vicinity of 1050 nm.

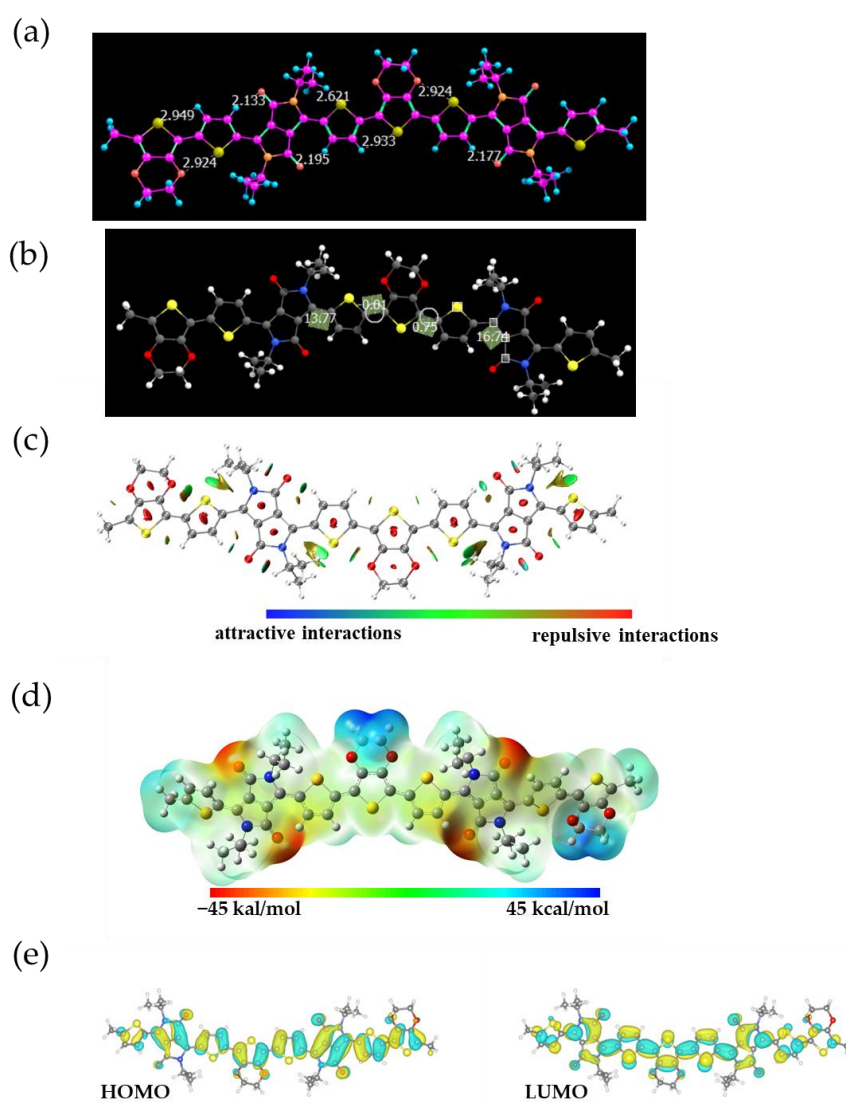


Figure 5. (a) Intramolecular distances and (b) dihedral angles of the dimer; (c) the HOMO and LUMO orbital energy diagram (d) RDG analysis and (e) ESP analysis of the isopropyl-substituted dimer.

2.5. OFET Device Performance

In order to determine the difference in conductivity between the two materials in terms of charge transfer, we fabricated bottom gate bottom contact (BGBC) structured OFET devices based on polymers P1 and P2. The structure of the devices is shown in Figure S5, and the preparation protocols of OFETs are placed in the Materials and Methods section. Devices based on P1 or P2 demonstrate only hole mobility but no electron mobility, which is typical of unipolar P-type organic semiconductor materials. Table 3 concludes the hole mobility extracted from the transfer characteristic curve (Figure 6a–b), and it can be seen that P2 outperforms P1 in terms of device performance. The devices with P2 have a saturation hole mobility about ten times higher than that of P1 and benefit from a high on/off current ratio (I_{on}/I_{off}).

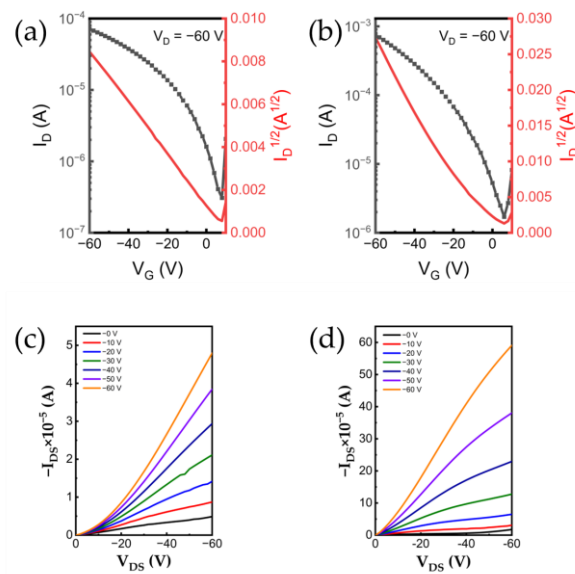


Figure 6. Conductivity transfer curves based on (a) P1 and (b) P2; the output curves based on the (c) P1 and (d) P2 device.

Table 3. Hole transport properties of polymer devices.

Polymer Material	Coating Speed (rpm)	Annealing Temperature (°C)	Max Hole Mobilities ¹ (cm ² /(V s))	Hole Mobilities ² (cm ² /(V s))	Threshold voltage (V)	Ion/Ioff
P1	2000	180	0.009	0.007	10	10 ²
P2	2000	180	0.15	0.12	-10	10 ³

¹ Measured under ambient conditions in saturation regime; ² Average of 6 devices tested.

2.6. Microstructure and Morphological Analysis of Polymer P1 and P2 Films

The microstructure of P1 and P2 in annealed films was investigated by atomic force microscopy (AFM). The annealed films of P1 and P2 show good crystallinity with no significant phase separation, which facilitates conductive hole migration. Figure 7a shows that the morphology of polymer P1 is a fibrous network structure with a small root-mean-square roughness (R_q) of 0.88 nm. The morphology of polymer P2 is not significantly different from that of P1, the only difference being that it has a larger R_q value of 1.35 nm. The ordered arrangement of the crystal domains is relatively more favorable to improve the electrical conductivity. The 3D topography of the polymer is placed in Figure S6.

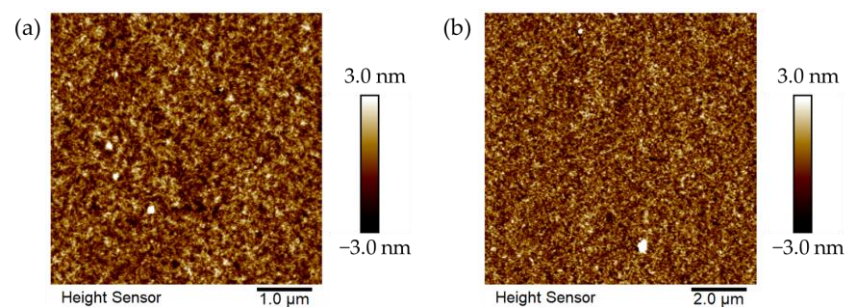


Figure 7. AFM height image of annealed film of (a) P1 and (b) P2.

3. Materials and Methods

Materials: Raw materials such as Pd₂(dba)₃ and tri-*o*-tolylphosphane (P(*o*-tol)₃) and organic solvents such as methanol, hexane etc. were purchased from Sigma-Aldrich (St. Louis, MO, USA). The dried chlorobenzene (water ≤ 50 ppm) is preserved through molecular 3A sieves. 5,7-bis(trimethylstannyl)-2,3-dihydrothieno [3,4-*b*][1,4]dioxine (EDOT-SnBu₃) was acquired from SunaTech (Suzhou, China). Both DPP-Br-C8C10 and DPP-Br-C10C12 are prepared from DPP units by alkylation and bromination reactions [49,50].

Polymerization method of P1: DPP-Br-C8C10 (210.0 mg, 0.2 mmol), EDOT-Sn (96.3 mg, 0.2 mmol), Pd₂(dba)₃ (3.7 mg, 4.0 μmol), and P(*o*-tol)₃ (5.0 mg, 16.0 μmol) were added to a Schlenk-tube under argon protection. Subsequently, anhydrous chlorobenzene solvent (11.0 mL) was added to the mixture. The reaction mixture was reacted at 130 °C and stirred for 12 h. The reaction was terminated and the mixture was gradually brought to room temperature and dropped into a beaker of methanol (200 mL) using a pipette. The powders are obtained by filtration and subsequently loaded into a Soxhlet extractor. Solvent extraction with methanol, acetone and hexane was carried out sequentially for 9 h to remove the oligomers. The extraction is followed by chloroform and the organic solution is concentrated and precipitated again dropwise in methanol (150 mL). The solids were filtered and then dried under vacuum (90 °C) to afford a black polymer (P1, 88% yield). The preparation of P2 was similar to that of P1 with the yield of 79%.

Characterization: The molecular weight of P1 and P2 were evaluated by the gel permeation chromatography (GPC, 150 °C, Agilent PL-GPC220, Atlanta, GA, USA). The eluent was trichlorobenzene to ensure adequate solubility. The solution at a concentration of 0.1 mg/mL was passed through the column (2* PLgel 10 μm MIXED-B) at a flow rate of 1.0 mL/min. Polystyrene and polystyrene-divinylbenzene were used as standards and as stationary phases, respectively. Thermal stability tests of P1 and P2 were conducted under nitrogen (TGA 550, Tainstruments, New Castle, DE, USA). Elemental analysis is measured by the CHN mode of organic elemental analyzer (Elementar Vario, Langenselbold, Germany). Photochemical characterization was performed on a UV-visible spectrometer (Cary 5000, Agilent, Santa Clara, CA, USA). The concentration of the polymer solution in chloroform solvent was around 0.5 mg/mL for testing in the solution state. The solution was further spin-coated onto a clean quartz plate (1.0 cm × 2.0 cm) and the solvent was evaporated by annealing to prepare a thin film for testing in the solid state. Electrochemical tests of the thin films were carried out in acetonitrile solutions containing tetrabutylammonium hexafluorophosphate as the electrolyte. Ag/AgCl electrode, glassy carbon electrode and platinum electrode were applied as reference electrodes, working electrodes and counter electrodes, respectively. A 1.0 mg/mL polymer chloroform solution was prepared and 6.0 μL of it was pipetted onto a glassy carbon electrode and allowed to evaporate slowly to form a dense film. The HOMO and LUMO energy levels were estimated by extracting data from the potentials at the onset of the oxidation peak and the onset of the reduction peak, respectively, according to the equations: $E_{\text{HOMO}} = -4.80 - (E_{\text{ox}}^{\text{onset}} - 0.40) \text{ eV}$; $E_{\text{LUMO}} = -4.80 - (E_{\text{red}}^{\text{onset}} - 0.40) \text{ eV}$, where E^{onset} is the potential value of the oxidation or reduction measured with respect to the Fc/Fc⁺ redox pair of the measured oxidation or reduction potential value. Thermally annealed films prepared under the same conditions as those used for OFET testing were measured with an atomic force microscope to observe their morphology (Nanoscope V, Bruker, Ettlingen, Germany).

Device preparation: Highly doped n-type silicon (Si) wafers with 300 nm silicon dioxide (SiO₂) content were used as substrates. The substrates were cleaned by ultrasonic cleaning in deionized water, acetone and isopropanol for 5 min, respectively. Octadecyltrichlorosilane (OTS) was used to modify the substrate. The gold was deposited as source and drain electrodes by photolithography. For the preparation of semiconductor films, P1 or P2 was pre-dissolved in chlorobenzene at a concentration of 6 mg/mL and heated overnight at 70 °C with stirring to ensure adequate solubility. The corresponding

polymer solutions were individually spin-coated onto the substrates at 2000 rpm (60 s) and then annealed in a glove box under nitrogen at 180 °C for 20 min.

The test of hole mobility of polymer-based OFET were characterized using the semiconductor 4200 characterization system (Keithley, USA) operating in the nitrogen glove box. The saturation hole mobility (μ) was calculated by the formula of: $\mu = \left(\frac{\partial \sqrt{|I_{DS}|}}{\partial V_{GS}}\right)^2 \cdot \frac{2L}{WC_i}$ (I_{DS} , V_{GS} , L and W are the source-drain current, gate voltage, channel length and channel width, respectively; C_i is the dielectric layer capacitance per unit area).

4. Discussion

This work introduces weak interaction forces through a molecular structure design strategy, which facilitates the planarity and structural regularity of the material. Suzuki polymerization conditions typically require the introduction of water to dissolve the base and facilitate the reaction, thus limiting the polymerization temperature and efficiency. Both P1 and P2 are fully polymerized at temperatures up to 130 °C in high yield, which allows for large volumes of such materials to be prepared, e.g., gram scale. Weak force-based strategies are often distinguished from covalent bond modulation of the molecular plane, which is more prominent for the stretching of materials. The attraction of the non-bonded form offers flexibility, facilitates recovery of the material after mechanical stress and offers the possibility of self-healing with resistance to tensile uplift. We believe that other materials designed based on the principle of intramolecular conformational locking could also provide inspiration for further exploration of device processing and preparation in organic electronics.

5. Conclusions

In this study, two polymers enduring different side chains were obtained by Stille coupling polymerization. Calculations based on theoretical simulations verify that the well coplanar nature of the materials is due to weak intramolecular forces. The side chains affect the weight-average molecular weight of the products and the catalytic efficiency, which is still unclear in terms of mechanism. The effect of different conjugation lengths on the thermal stability and the energy levels of the frontier orbitals of the materials is insignificant. On the other hand, different lengths of the side chains lead to variations in the stacking of the material from chain to chain, which causes a significant difference in its absorption in the film state. Conductivity tests based on organic transistors show significantly better hole mobility for polymer P2 compared to P1. Our study explores the effect of side chains on the material properties from a microscopic point of view, which aids in the further design of suitable polymer semiconductor materials. Research work on stretchable organic transistors and sensors for this range of π -conjugated polymers is still ongoing.

Supplementary Materials: The following supporting information can be downloaded at: www.mdpi.com/xxx/s1.

Author Contributions: Conceptualization, J.C. and A.Y.; methodology, S.R.; software, W.Z.; validation, S.R., J.C. and A.Y.; formal analysis, S.R.; investigation, W.Z.; resources, W.Z.; data curation, S.R.; writing—original draft preparation, S.R.; writing—review and editing, J.C.; visualization, A.Y.; supervision, J.C.; project administration, A.Y.; funding acquisition, S.W. and J.C. All authors have read and agreed to the published version of the manuscript.

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