



Role of polymer side chains in dopant diffusion into conjugated polymer films during solution sequential doping

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Abstract

The role of polymer side chains in dopant diffusion during solution sequential doping is systematically investigated using poly(3–2-methyl-2-hexylcarboxylate)thiophene (P3ET), a polythiophene derivative with thermocleavable side chains. Two types of polymer films, P3ET with intact side chains and side-chain-free P3ET-COOH obtained by thermocleavage, are subjected to sequential treatment with an acetonitrile solution of the benchmark molecular dopant 2,3,5,6-tetrafluoro-7,7,8,8-tetracyanoquinodimethane (F4TCNQ) and compared. Spectroscopic and microstructural analyses reveal that although comparable total amounts of F4TCNQ are incorporated into both films, their spatial distribution and aggregation behavior differ markedly. In P3ET-COOH, F4TCNQ is strongly concentrated and crystallized on the film surface, whereas in P3ET it is less concentrated and only weakly crystallized. The difference is attributed to restricted F4TCNQ diffusion in the P3ET-COOH film, arising from the absence of side chains and minimized swelling. These findings highlight the critical role of polymer side chains in enabling efficient and uniform doping of conjugated polymers via solution sequential doping.

Yongkyeong Lee and Juyeong Bae have contributed equally to this work.

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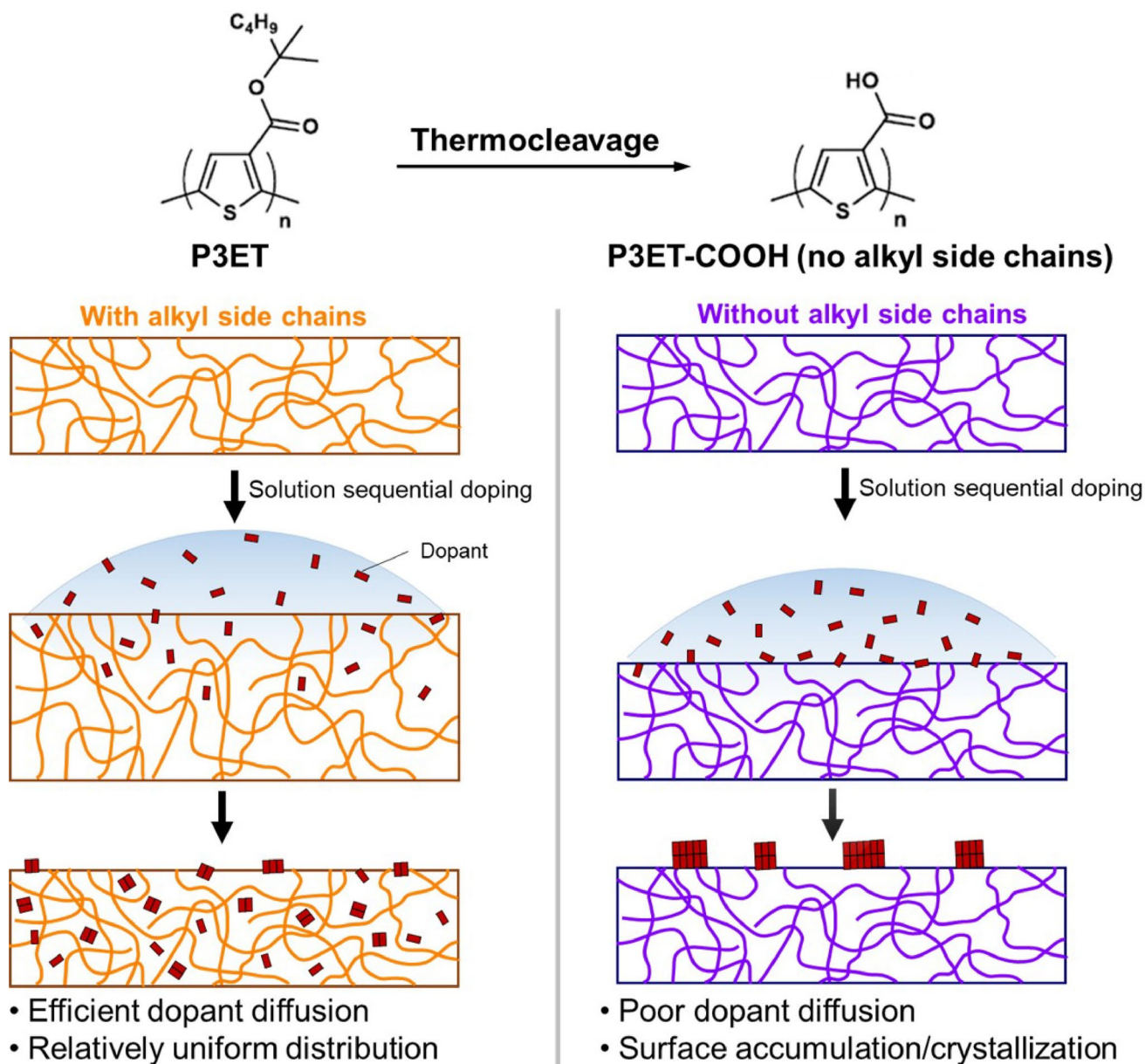
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Graphical abstract

This study reveals how polymer side chains govern dopant diffusion in conjugated polymer films during solution sequential doping. By comparing a conjugated polymer with its side-chain-free derivative, this work shows that side chains are essential for efficient dopant penetration and uniform distribution. In their absence, dopant molecules localize on the film surface due to severely restricted diffusion.



Keywords Conjugated polymers · Molecular dopants · Side chains · Solution sequential doping · Thermocleavage

1 Introduction

Doping of conjugated polymers is widely utilized and actively studied across various areas of organic electronics, including organic thermoelectric devices [1–9], organic transistors [10–14], organic solar cells [15–18],

and polymer electrodes [19–22]. Among the different processing methods, solution sequential doping involves first forming a solid polymer film, followed by applying a dopant solution prepared in a solvent that does not dissolve the polymer but readily dissolves the dopant molecules [23]. The dopant solution is left in contact with the polymer film

for a defined period, allowing dopants to be incorporated. During this sequential treatment, dopant molecules diffuse from the film surface into its bulk, a process accompanied by charge transfer between the dopant and the polymer [24]. This charge transfer generates mobile charge carriers within the polymer, thereby achieving doping.

Previous studies have demonstrated that, in solution sequential doping, the interaction between the polymer side chains and the dopant solution plays a critical role in determining the doping efficiency and the electrical properties of the resulting doped films [25–30]. For example, S. E. Yoon et al. reported that the choice of dopant solvent significantly influences the doping efficiency of solution sequential doping [26]. They showed that high doping efficiency can be achieved when the dopant solvent exhibits both high solubility for the polymer and sufficient polarity to dissolve the dopant, highlighting the importance of the solvent's ability to solubilize polymer side chains. More recently, J. Kim et al. explicitly demonstrated the critical role of side chain-solvent interactions in the solution sequential doping of conjugated polymers [28]. In their study, the doping of poly[2,5-bis(3-tetradecylthiophen-2-yl)thieno[3,2-b]thiophene] (PBTTT) with tris(pentafluorophenyl)borane (BCF) was highly inefficient when acetonitrile (ACN), a polar solvent, was used, but effective when hexane, a non-polar aliphatic solvent, was employed. This contrast was attributed to the much stronger interaction between hexane and the side chains of PBTTT compared to ACN. Similarly, V. Vijayakumar et al. showed that in the solution sequential doping of PBTTT with 2,3,5,6-tetrafluoro-7,7,8,8-tetracyanoquinodimethane (F4TCNQ), both the doping kinetics and the final doping level strongly depend on the side chain length of PBTTT. They further demonstrated that side chains that are either excessively disordered or excessively crystalline hinder the effective diffusion of dopants within the polymer film [25].

As such, prior studies have clearly established the critical role of polymer side chains in the solution sequential doping of conjugated polymer films. However, the extreme case of conjugated polymers lacking side chains has received little attention to date. Investigating solution sequential doping in side chain-free conjugated polymers is expected to provide the most direct and pronounced insight into how side chains influence dopant diffusion during the doping process. Moreover, with recent reports identifying the doping of side chain-free conjugated polymers, or the post-doping removal of side chains, as promising strategies to achieve high electrical conductivity and improved doping stability [31–35], elucidating the solution sequential doping behavior of side chain-free conjugated polymer films is anticipated to support the development of such high-performance, stable doped materials. Although some studies have reported

improvements in the electrical conductivity of side chain-free conjugated polymer films through solution sequential doping [34, 35], little has been reported on the diffusion behavior of dopants within these films during the sequential doping process or on the spatial distribution of dopants in the resulting doped films.

In this context, this study investigates the role of polymer side chains in governing dopant diffusion during the solution sequential doping of conjugated polymer thin films, with particular attention to whether this method remains effective for polymers without side chains. Here, we note that the term “doping” is used in a general sense to refer to the physical incorporation of dopant molecules into the polymer film, even in cases where no actual charge transfer occurs between the dopant molecules and the polymer. Using P3ET, a polythiophene-based p-type conjugated polymer bearing thermocleavable side chains, we performed sequential treatment with a solution of the representative molecular p-dopant F4TCNQ on two types of films: one with intact side chains (P3ET) and one with side chains removed through thermal cleavage (P3ET-COOH). The results revealed a pronounced difference in the spatial distribution of F4TCNQ depending on the presence of side chains, despite nearly identical total uptake of F4TCNQ in both films. Specifically, in the absence of side chains, a much larger fraction of F4TCNQ remained localized at the film surface, where severe crystallization of F4TCNQ molecules occurred. This observation indicates that the lack of side chains strongly restricted the diffusion of F4TCNQ into the polymer bulk. We attributed this inhibited diffusion to the absence of favorable side chain-solvent interactions and the minimal swelling of side chain-free films, both of which are likely to suppress the diffusivity of F4TCNQ within the film.

2 Experimental

2.1 Materials

Poly(3-(2-methyl-2-hexylcarboxylate)thiophene) (P3ET), a polymer obtained from the polymerization of a thiophene monomer bearing a thermocleavable tertiary ester side chain, was synthesized via chain-growth Suzuki polycondensation, as we have previously reported in detail in our earlier publication [36, 37]. F4TCNQ (purity > 98%), was obtained from Tokyo Chemical Industry Co., Ltd. (Japan). Acetonitrile (ACN, ≥ 99.5%) and anhydrous chloroform (CF, ≥ 99%) were supplied by Sigma-Aldrich Inc. (Germany). F4TCNQ, ACN, and CF were employed as received, without additional purification.

2.2 Preparation of polymer films

P3ET was dissolved in CF at 55 °C to prepare a 5 mg mL⁻¹ solution. The P3ET solution was spin-coated onto substrates (glass or silicon wafers with a native oxide layer) at 2000 rpm for 30 s to form thin films. The resulting P3ET films were dried at room temperature for 1 h, followed by thermal treatment on a hot plate at 200 °C for 10 min to remove the side chains via thermocleavage. The polymer film with side chains removed through thermal treatment will hereafter be referred to as P3ET-COOH. After heating, the P3ET-COOH films were cooled to room temperature for 10 min. All procedures were carried out in a nitrogen-filled glovebox. Prior to polymer film deposition, the substrates were cleaned by sequential ultrasonication in detergent solution, deionized water, acetone, isopropyl alcohol, and deionized water. The cleaned substrates were then thoroughly dried and treated with UV–ozone for 20 min to improve the wetting of the polymer solution on the surface.

2.3 Treatment of polymer films with F4TCNQ solution

P3ET and P3ET-COOH films were treated with an F4TCNQ solution following the standard solution sequential doping procedure. An F4TCNQ solution with a concentration of 2 mg mL⁻¹ was prepared by dissolving F4TCNQ in ACN at room temperature. This solution was dropped onto the P3ET or P3ET-COOH film to fully cover its surface and left for 1 min to allow the F4TCNQ to diffuse into the polymer film. The excess F4TCNQ solution on the surface was then removed by spinning the sample in a manner similar to spin coating, after which the films were thoroughly dried. All procedures were carried out inside a nitrogen-filled glovebox.

2.4 Characterizations

Fourier transform infrared (FT-IR) spectroscopy was performed using a VERTEX 7-v FT-IR spectrometer (Bruker), and ultraviolet–visible–near infrared (UV–Vis–NIR) absorption spectroscopy was conducted using a V-770 spectrometer (Jasco). Grazing-incidence X-ray diffraction (GIXD) measurements were carried out at the 3C and 9A

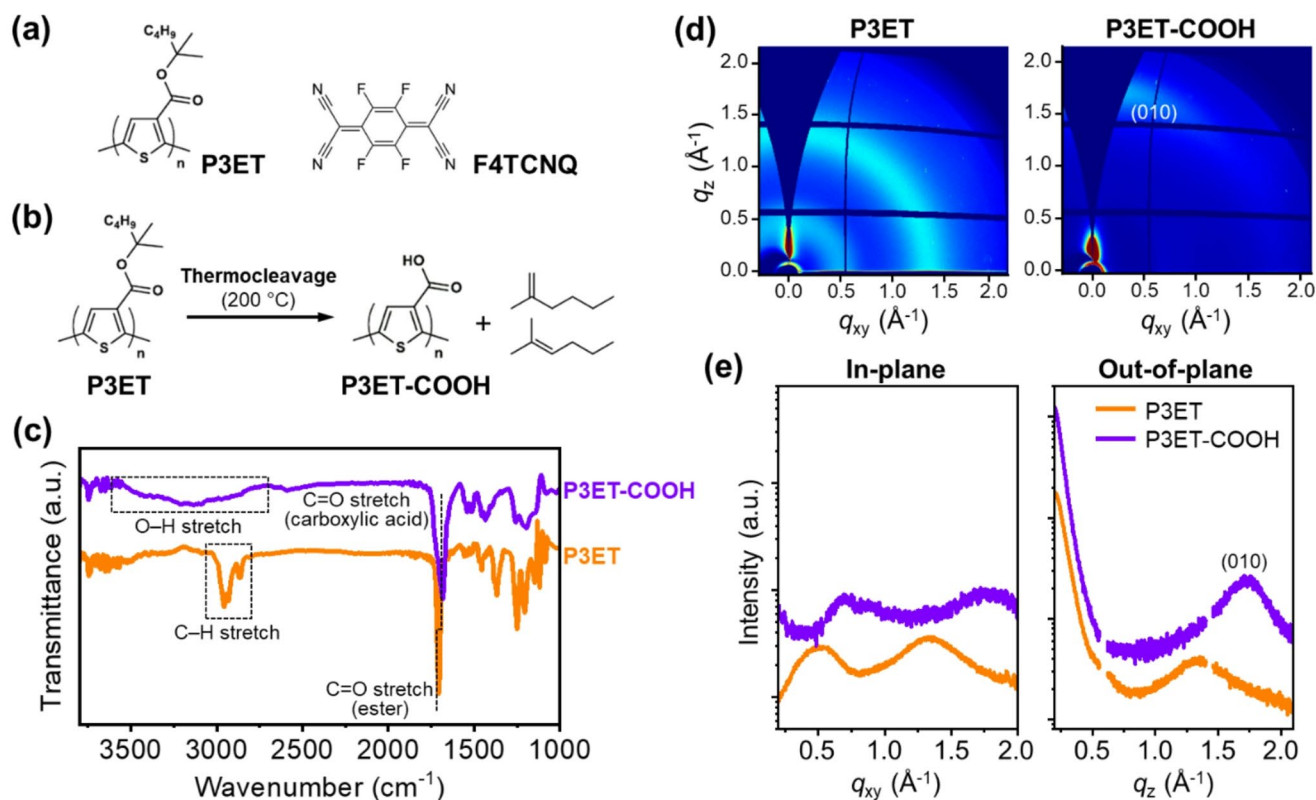


Fig. 1 Materials used in this study and their characterization. **a** Molecular structures of P3ET and F4TCNQ. **b** Thermocleavage of the alkyl side chains of P3ET upon thermal treatment, leading to the formation of P3ET-COOH. **c** FT-IR spectra of P3ET and P3ET-COOH films. **d**

Two-dimensional diffraction patterns obtained from GIXD measurements of P3ET and P3ET-COOH films. **e** Line-cut intensity profiles of the diffraction patterns shown in **d**, taken along the in-plane (left) and out-of-plane (right) directions

beamlines of the Pohang Accelerator Laboratory (PAL), with an X-ray wavelength of 1.33 Å and a sample-to-detector distance of 214 mm. The incidence angle of the X-ray was set to the critical angle of the samples. X-ray photoelectron spectroscopy (XPS) was performed using a NEXSA G2 system (Thermo Fisher Scientific). The thickness of the polymer films was measured using an M-2000 ellipsometer (J.A. Woollam) by fitting the spectra in the wavelength range of 850–1000 nm with the Cauchy model. To measure the thickness of polymer films swollen by ACN, the samples were immersed in pure ACN for 1 min, the surface liquid was quickly removed by nitrogen blowing, and ellipsometry was immediately performed.

3 Results and discussion

Figure 1a shows the molecular structures of P3ET, the conjugated polymer investigated in this study, and F4TCNQ, a representative molecular dopant commonly employed for p-doping of conjugated polymers. P3ET consists of a polythiophene backbone with tertiary ester side chains that undergo thermocleavage at approximately 180–200 °C, leaving short carboxyl groups after removal of the alkyl chains. The thermocleavage process is illustrated in Fig. 1b, and its detailed mechanism has been reported previously [36, 38]. Note that at this temperature, the polymer exhibited no observable thermal degradation of the conjugated backbone other than the thermocleavage of its side chains [36, 37]. By exploiting the thermocleavable nature of the tertiary ester groups, we removed the long alkyl side chains from as-cast P3ET films through thermal treatment. Specifically, P3ET films were spin-coated from solution and annealed at 200 °C for 10 min on a hot plate, yielding P3ET-COOH films in which most alkyl chains were cleaved. The full preparation procedure is described in the Experimental section. Consequently, two types of films with identical polythiophene backbones but different side chain content were obtained: P3ET (with side chains) and P3ET-COOH (without side chains).

Thermocleavage of the side chains was confirmed by FT-IR spectroscopy (Fig. 1c). Prior to thermal annealing, the P3ET film displayed peaks at 2800–3000 cm^{-1} corresponding to C–H stretching vibrations of alkyl side chains. After thermal annealing, these peaks disappeared, and a broad O–H stretching band appeared at 2700–3300 cm^{-1} . In addition, the C=O stretching peak at $\sim 1706 \text{ cm}^{-1}$ (ester) shifted to $\sim 1685 \text{ cm}^{-1}$ (carboxylic acid), further confirming side chain cleavage [36]. These changes clearly indicate that thermal treatment effectively converted P3ET to P3ET-COOH.

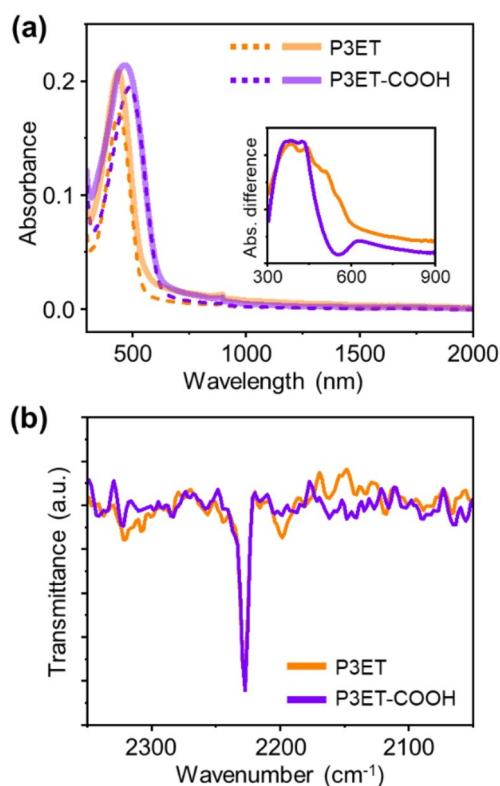


Fig. 2 Spectroscopic analysis of P3ET and P3ET-COOH films after sequential treatment with F4TCNQ solution. **a** UV-vis-NIR absorption spectra of each polymer film before (dotted line) and after (solid line) sequential treatment. Inset: difference spectrum obtained by subtracting the pre-treatment spectrum from the post-treatment spectrum. **b** FT-IR spectra of each polymer film after sequential treatment

Structural differences between the films were also evident in GIXD patterns (Fig. 1d and e). The P3ET film exhibited no discernible diffraction peaks, consistent with its near-amorphous character arising from bulky ester side chains that hinder molecular packing. In contrast, the P3ET-COOH film displayed a (010) diffraction peak at $q \sim 1.7 \text{ Å}^{-1}$ in the out-of-plane direction, indicative of π - π stacking facilitated by reduced steric hindrance. These results are consistent with previous reports [37].

Sequential treatment with an F4TCNQ solution was then performed on both P3ET and P3ET-COOH films. F4TCNQ was dissolved in ACN, an orthogonal solvent for the polymers and a widely medium for solution sequential doping of conjugated polymers. The full procedure is described in the Experimental section. Notably, we deliberately use the term “sequential treatment” rather than “sequential doping.” This distinction reflects the fact that no electron transfer occurs between F4TCNQ and either P3ET or P3ET-COOH, as demonstrated later. The absence of electron transfer arises from the twisted conformation of the polythiophene backbone induced by ester and carboxyl groups, which lowers the HOMO energy levels of P3ET and P3ET-COOH

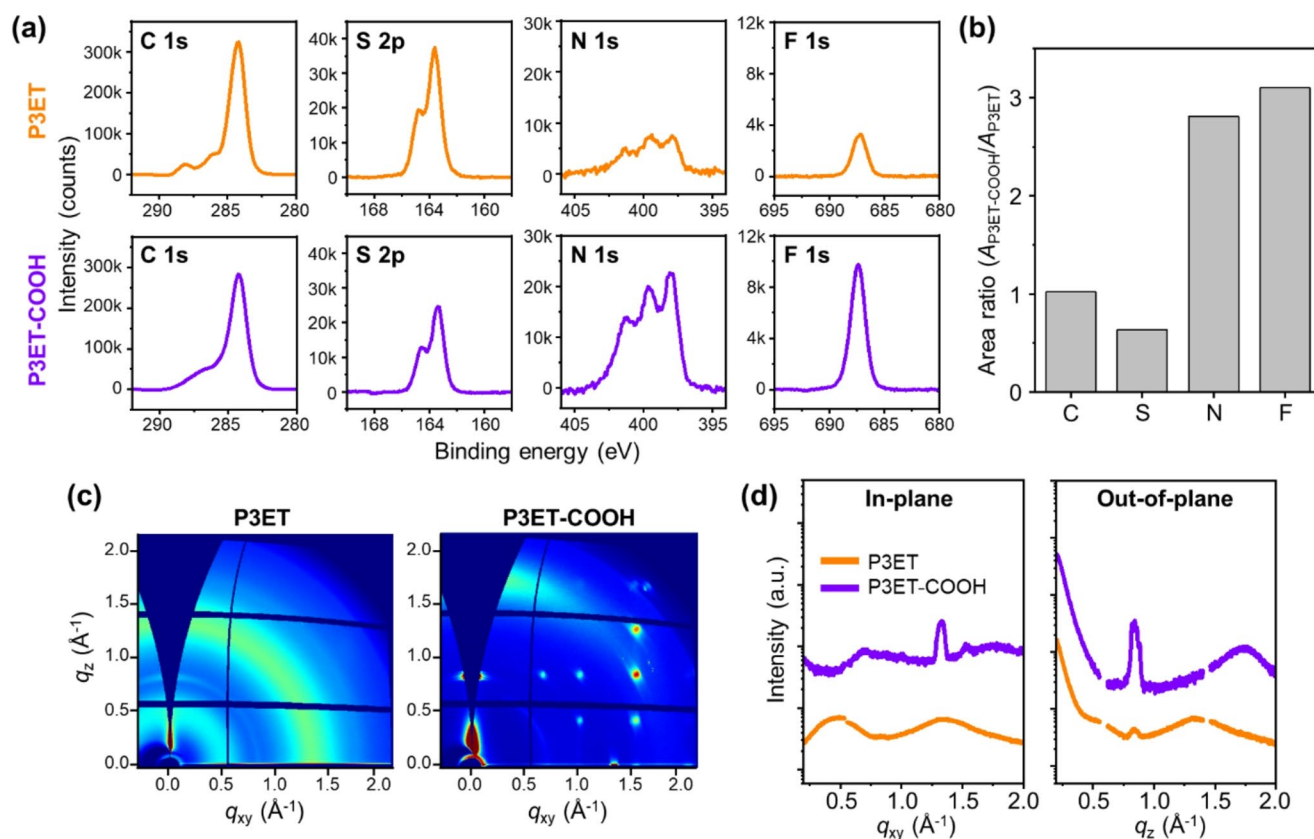


Fig. 3 XPS and GIXD analyses of P3ET and P3ET-COOH films after sequential treatment with F4TCNQ solution. **a** XPS spectra (C 1 s, S 2p, N 1 s, and F 1 s, from left to right) of the film surfaces. **b** Relative elemental contents at the film surfaces, calculated from the spectra in **a**. Ratios are shown as the peak areas of P3ET-COOH normalized

to those of P3ET. **c** Two-dimensional GIXD diffraction patterns of sequentially treated P3ET and P3ET-COOH films. **d** Line-cut intensity profiles of the diffraction patterns in **c** along the in-plane (left) and out-of-plane (right) directions

far below the LUMO of F4TCNQ [34]. This unfavorable alignment of energy levels prevents electron transfer from the polymer HOMO to the F4TCNQ LUMO. Although electron transfer is absent in this system, it remains highly suitable for our purpose: comparing dopant diffusion in polymers with and without side chains. In fact, the lack of electron transfer enables us to isolate the role of physical diffusion, without complications from Coulombic interactions between polymer polarons and F4TCNQ anions. We acknowledge that diffusion behavior could differ in systems where electron transfer occurs [24], and we consider comparative studies of such cases to be an important subject for future investigation.

After performing sequential F4TCNQ solution treatment under identical conditions on P3ET and P3ET-COOH films, we confirmed that the total amount of F4TCNQ incorporated into the films was nearly identical, as verified by UV-vis-NIR and FT-IR measurements (Fig. 2). Figure 2a shows the UV-vis-NIR absorption spectra of P3ET and P3ET-COOH before and after treatment. The pristine P3ET and P3ET-COOH films exhibited maximum absorption at

440 nm and 485 nm, respectively, without any shoulder peaks, indicating that intermolecular packing between polymer chains was not significant. This observation is consistent with the GIXD results shown in Figs. 1d and e. The absorption edges were located at 535 nm for P3ET and 615 nm for P3ET-COOH. The red-shift in the absorption of P3ET-COOH relative to that of P3ET suggests an extended effective conjugation length of P3ET-COOH, which can be attributed to the enhanced planarity of the polymer backbone after removal of side chains by thermocleavage, as supported by the GIXD analysis. After sequential F4TCNQ solution treatment, the absorption increased slightly in the range of 300–600 nm. No polaronic absorption band appeared in the near-IR region, confirming that no charge transfer occurred between F4TCNQ and the polymers [39], as previously discussed. The difference spectra, obtained by subtracting the pre-treatment spectra from the post-treatment spectra, closely matched the reported absorption of neutral F4TCNQ in the literature [40–42]. This finding demonstrates that the observed spectral changes originate from the incorporation of neutral F4TCNQ molecules into

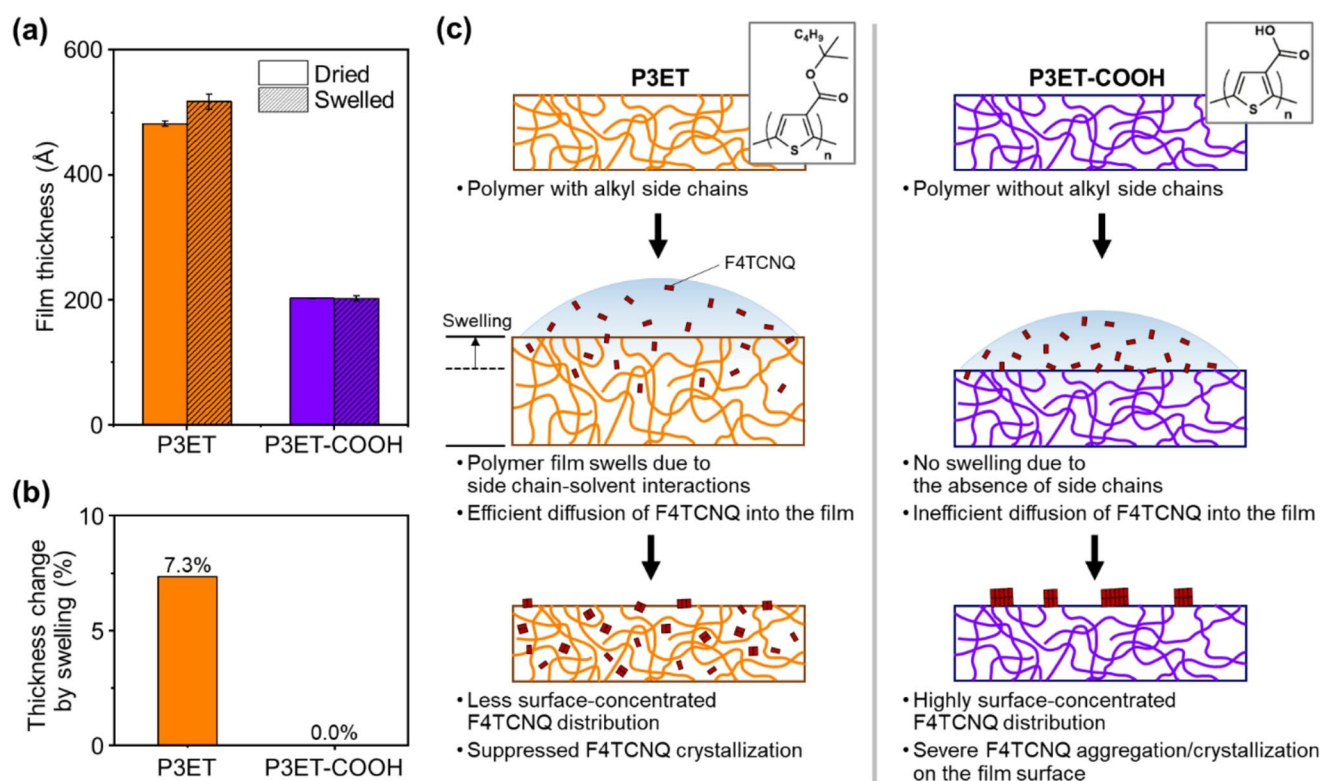


Fig. 4 Swelling behavior of P3ET and P3ET-COOH films and its relation to F4TCNQ diffusion. **a** Thickness of P3ET and P3ET-COOH films before and after immersion in pure ACN. **b** Percentage variation in thickness, calculated by normalizing the thickness change in **a** to the

initial thickness before swelling. **c** Schematic illustration showing how the presence (P3ET) or absence (P3ET-COOH) of alkyl side chains influences polymer film swelling, F4TCNQ diffusion, and the distribution of F4TCNQ in the final films after sequential treatment

the polymer films. Furthermore, the maximum absorption intensity in the difference spectra was nearly identical for P3ET and P3ET-COOH, indicating that the incorporated amount of F4TCNQ was essentially the same in both cases.

Consistent results were obtained from FT-IR analysis (Fig. 2b). Both P3ET and P3ET-COOH films exhibited the $\text{C}\equiv\text{N}$ stretching vibration of neutral F4TCNQ at 2227 cm^{-1} [43], with nearly identical intensities. These results further confirm that comparable amounts of neutral F4TCNQ were incorporated into the two types of polymer films.

We next performed XPS analysis to quantify the amount of F4TCNQ present on the surface of each sequentially treated polymer film (Figs. 3a and b). The integrated area of the C 1s peak was similar for both P3ET and P3ET-COOH. However, despite sharing the same polythiophene backbone, the S 2p peak area was smaller for P3ET-COOH than for P3ET. In contrast, the N 1s and F 1s peak areas were approximately three times larger for P3ET-COOH, indicating that the surface concentration of F4TCNQ in P3ET-COOH films (within the XPS sampling depth) was roughly three times greater than in P3ET films. The reduced S 2p peak intensity for P3ET-COOH can be attributed to the higher surface coverage of F4TCNQ, which diminishes the relative signal contribution from the polymer backbone.

GIXD measurements further revealed distinct differences in the crystalline characteristics of F4TCNQ on the treated films (Figs. 3c and d). In P3ET, the F4TCNQ solution treatment resulted only in the appearance of a faint circular pattern at $q \sim 0.84\text{ Å}^{-1}$ [44], suggesting weak crystallization with random crystalline orientation. By contrast, the P3ET-COOH film exhibited intense and well-defined diffraction peaks after treatment, matching the features of pristine crystalline F4TCNQ [43]. These results indicate that while F4TCNQ crystallizes only weakly and isotropically in the P3ET film, crystallization of F4TCNQ occurs only weakly and isotropically in P3ET, it crystallizes strongly in P3ET-COOH, forming larger and more oriented crystals.

Meanwhile, the GIXD results provide additional indirect experimental evidence for the limited incorporation of F4TCNQ into the P3ET-COOH film, as indicated by the unchanged position of its (010) peak before and after the sequential F4TCNQ solution treatment. It is well established that when F4TCNQ is incorporated into a conjugated polymer film, the crystalline structure of the polymer is modified, typically resulting in a decreased π - π stacking distance [6, 8]. However, in the case of P3ET-COOH, the position of the (010) diffraction peak remains unchanged at $q = 1.72\text{ Å}^{-1}$ before and after F4TCNQ treatment (Figs. 1e

and d), indicating that the π – π stacking distance is unaffected by the treatment process. This observation provides indirect evidence that the amount of F4TCNQ incorporated into the P3ET-COOH film is minimal.

Taken together, the XPS data show a significantly larger amount of F4TCNQ localized at the surface of P3ET-COOH, and the GIXD results demonstrate that this high surface concentration promotes robust crystallization of F4TCNQ. Considering that the total amount of incorporated F4TCNQ is nearly identical for both polymers, as shown earlier, it can be inferred that in P3ET the dopant is relatively uniformly distributed throughout the bulk, whereas in P3ET-COOH it is preferentially concentrated at the surface, enabling pronounced crystallization.

The above results clearly demonstrate the critical role of polymer side chains in governing the diffusion of F4TCNQ into the polymer films during solution sequential treatment. In the absence of side chains, F4TCNQ diffusion is severely restricted, resulting in inefficient penetration into the film interior. Consequently, F4TCNQ accumulates at the film surface, where strong aggregation and crystallization occur. This behavior can be attributed primarily to the lack of film swelling when side chains are absent, which limits the mobility of polymer chains and solvent molecules within the film [45, 46]. Indeed, swelling measurements using ACN confirmed this interpretation (Fig. 4a and b): while pristine P3ET expanded from an initial thickness of 48.2 nm to 51.7 nm after ACN exposure, corresponding to a 7.3% increase, P3ET-COOH maintained a constant thickness of about 20.2 nm before and after ACN exposure, indicating negligible swelling. A schematic illustration that correlates the molecular structure of the polymers with their swelling behavior, F4TCNQ diffusion behavior, and the resulting F4TCNQ distribution in the final films, summarizing all the observations made thus far, is presented in Fig. 4c.

Finally, we note that the dopant distribution within the polymer films, which varies depending on the presence or absence of side chains as observed in this study, may be influenced by the molecular size of the dopant used. It has been reported that smaller dopant molecules possess higher diffusivity in polymer matrices and therefore tend to penetrate deeper into the film, whereas larger dopant molecules are less mobile and exhibit much slower diffusion rates [47]. Accordingly, we expect that if a dopant molecule smaller than F4TCNQ were used in this study, the difference in dopant distribution between P3ET and P3ET-COOH would become less pronounced, as the smaller dopant could more easily migrate into the side chain-free polymer matrix. Conversely, the use of a bulkier dopant would likely amplify this contrast. We believe that systematically investigating how the presence or absence of polymer side chains affects

dopant diffusion as a function of dopant molecular size would be an interesting topic for future study.

4 Conclusions

In summary, we investigated the influence of polymer side chains on dopant diffusion into polymer films during solution sequential treatment, using P3ET as a model conjugated polymer whose side chains can be almost completely removed by thermocleavage. A comparison between P3ET with intact side chains and side chain-free P3ET-COOH revealed that, although both polymers incorporated a comparable total amount of F4TCNQ upon treatment, their surface distribution and crystallization behavior differed markedly. In P3ET films, F4TCNQ showed relatively mild surface accumulation and weak, randomly oriented crystallization. By contrast, in P3ET-COOH films, approximately three times more F4TCNQ accumulated at the surface, accompanied by strong crystallization with preferential orientation. This pronounced surface localization and enhanced crystallization of F4TCNQ in P3ET-COOH arise from inefficient diffusion of F4TCNQ into the film bulk caused by the absence of side chains. These findings highlight the critical role of side chains in enabling effective dopant diffusion during solution sequential doping. Moreover, they suggest that achieving uniform doping and high doping levels in side chain-free conjugated polymer films may require alternative approaches beyond solution sequential treatment.

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Author contributions YL, JB: conceptualization, methodology, formal analysis, data curation, writing—original draft, writing—review and editing. SYS: supervision, methodology. HL: supervision, methodology, funding acquisition, writing—original draft, writing—review and editing.

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Declarations

Conflict of interest The authors declare no competing interests.

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