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Research Progress on Intrinsic Material Strategies for D-A Conjugated Polymers in Stretchable Polymer Optoelectronic Devices

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Abstract. D-A conjugated polymers play a pivotal role in stretchable optoelectronic devices due to their tunable energy levels and high charge mobility, finding widespread application in flexible organic electronics such as field-effect transistors, solar cells, and wearable sensors. These devices must maintain robust optoelectronic functionality under complex mechanical deformation, with performance highly dependent on the material's intrinsic mechanical adaptability. Unlike brittle inorganic materials, the molecular architecture of D-A polymers positions them as pivotal for advancing stretchable technologies. This paper focuses on the application progress of D-A conjugated polymers in stretchable optoelectronic devices, particularly exploring how molecular design achieves synergistic optimization between optoelectronic performance and mechanical flexibility. Through an in-depth analysis of material design strategies, this paper summarizes current research achievements in enhancing material mechanical adaptability and optoelectronic performance at the molecular level, proposing design concepts for high-performance, intrinsically stretchable polymer optoelectronic materials. Finally, it briefly reviews research progress and challenges in related fields, while outlining future directions for the design and application of high-performance stretchable polymer optoelectronic materials.

Keywords: D-A conjugated polymers, optoelectronic materials, polymer semiconductors, stretchable polymer optoelectronic devices

1. Introduction

With the rapid advancement of flexible electronics, stretchable optoelectronic devices have demonstrated revolutionary potential in fields such as health monitoring, electronic skin, and human-machine interaction. Unlike traditional rigid devices, these flexible components must maintain stable electrical signal generation and transmission capabilities under repeated multi-axis mechanical deformations, including bending, twisting, and stretching. This necessitates mechanical flexibility across all constituent materials. Among them, the organic functional layer of the semiconductor—the core for charge transport—must strike a balance between high electrical performance (such as high mobility, on/off ratio, and low threshold voltage) and high mechanical

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flexibility (such as high relative elongation and low elastic modulus). Traditional silicon-based or metal oxide semiconductor materials, due to their inherent brittleness, are prone to cracking and failure under large strains, making them unsuitable for these demands. Consequently, material design must be re-examined to develop stretchable, high-performance materials for all components of flexible electronics. Organic semiconductors stand out due to their molecular designability and solution processability. Among these, D-A conjugated polymers, owing to their unique electronic structure, have emerged as a key material system for advancing stretchable technology.

D-A conjugated polymers achieve high charge mobility and tunable optoelectronic properties through the precise alternating arrangement of strong electron-donating and electron-accepting units, which induces intramolecular charge transfer effects and regulates the polymer's energy levels and bandgap. They are thus regarded as ideal candidates for intrinsic stretchable semiconductors [1, 2]. However, D-A conjugated backbones typically exhibit highly conjugated planar structures with elevated crystallinity, rendering the material inherently rigid and resistant to stretching. This mechanical fragility means that even if other components such as electrodes or dielectrics possess elasticity, any cracking or discontinuity in the semiconductor layer leads to a drastic degradation in device performance. This severely limits their practicality in applications such as human motion monitoring and implantable medical devices. Consequently, endowing conjugated polymers with both high electrical performance and mechanical extensibility has emerged as a major challenge in flexible electronics [3].

To address this bottleneck, recent research has focused on intrinsic material strategies. Through molecular-scale design, these approaches confer high extensibility and mechanical stability upon polymers without sacrificing optoelectronic properties [4]. Compared to achieving stretchability through structural design, intrinsic material strategies aim for molecular-level engineering. These approaches primarily encompass two directions: backbone engineering and side-chain engineering. Backbone engineering optimizes backbone flexibility and charge transport pathways through configuration regulation, dynamic bond introduction, and topological decoupling. Side-chain engineering balances crystallization behavior and intermolecular interactions by utilizing flexible units, branched structures, and anchor positioning. Against this backdrop, this article reviews recent advances in the field of internal material strategy to provide a theoretical basis and practical recommendations for the design of materials for high-performance polymer optoelectronic devices.

2. Main-chain engineering molecular design

Main-chain engineering aims to enhance the deformation compliance of polymer backbones during stretching through molecular structural regulation of the conjugated framework, while maintaining or improving their optoelectronic properties. D-A polymers typically exhibit highly coplanar and crystalline main chains, which favour charge carrier transport but compromise mechanical extensibility. Consequently, main chain engineering seeks to introduce structural modifications that reduce chain rigidity or enhance deformability without disrupting conjugation continuity. This section outlines three intrinsic material strategies at the main chain level to explore their rationale.

2.1. Configuration control strategy

Traditional linear conjugated polymers exhibit high backbone rigidity and crystallization tendency, often initiating microcracks during stretching. Configuration control strategies can modulate local backbone rigidity or flexibility through single-bond rotational steric hindrance, non-covalent conformational locks, or locally twisted unit design, without disrupting the conjugated backbone

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continuity [5]. Two primary approaches exist: the first involves embedding twisted units with specific geometric configurations within the conjugated backbone to serve as forced kink points. Zhu et al. constructed a series of ortho/ o-substituted D-A copolymers, introducing ortho- or meta-substituted biphenyl units at 5% or 10% by weight into a DPP-(bithiophene) backbone. This transformed the linear conjugated chain into a series of zigzag-structured polymers [6]. One variant, designated 'o-OC8-5%', exhibited outstanding properties, as illustrated in Figure 1. Its organic field-effect transistors (OFETs) exhibited an initial hole mobility as high as 1.92 $cm^2/V \cdot s$. This mobility remained at 1.43 $cm^2/V \cdot s$ when stretched to 100% strain parallel to the transport direction and at 1.37 $cm^2/V \cdot s$ when stretched perpendicular to the transport direction [6]. Furthermore, the material demonstrated negligible performance degradation after multiple stretching cycles, reflecting outstanding mechanical stability. This molecular design absorbs tensile strain through conformational transitions while preserving long-range π-π stacking and short-range random distortion, achieving dual enhancement of both mobility and stretchability.

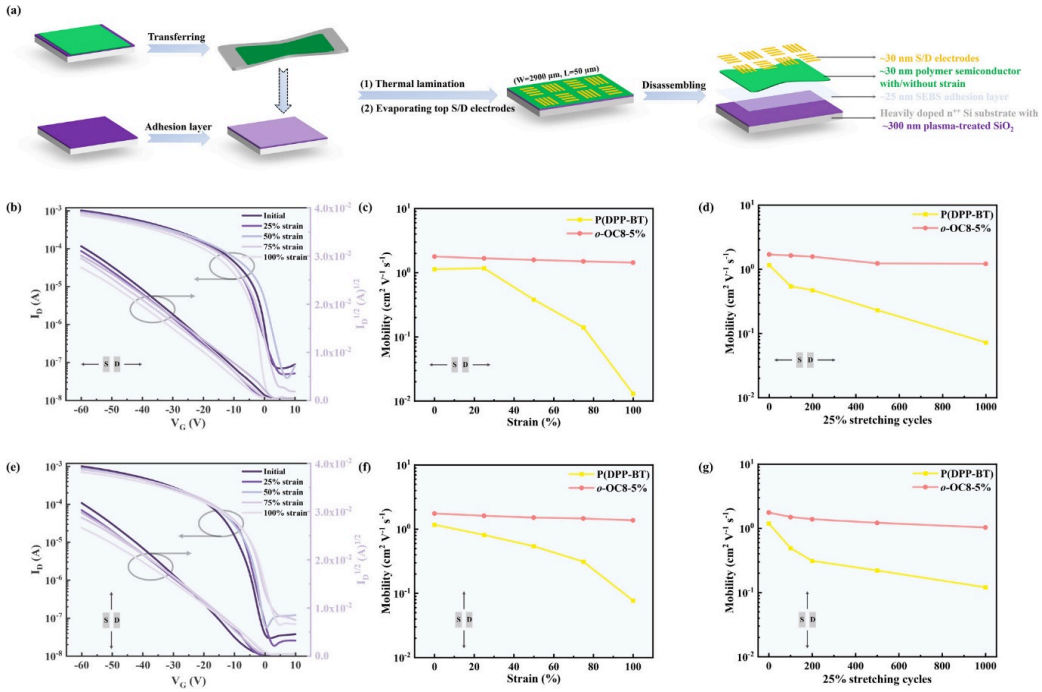

Figure 1. Molecular-scale geometric design: zigzag-structured intrinsically stretchable polymer semiconductors. (a) Fabrication of BGTC-type FETs with SEBS layer. (b,e) Transfer curves of o-OC8–5% under increasing strain, with transport parallel/perpendicular to strain. (c,d,f,g) Mobilities of P(DPP-BT) and o-OC8–5% under varied strain and repeated stretching cycles [6]

Another approach involves disrupting the coplanarity of D-A conjugated polymers through bending points, such as the atomic substitution of the thiazole ring (N replacing C-H) and S···O non-covalent locking employed by Wu et al. [7]. This transforms the inherently flexible, easily distorted connecting region into a quasi-rigid structure while retaining the alkoxy group's solubility-enhancing properties [7]. This design overcomes the traditional dichotomy in functional partitioning where rigid domains solely handle transport and flexible domains solely accommodate deformation. It simultaneously achieves enhanced electron mobility (5.4×10^{-4} $cm^2/V \cdot s$), tighter π-π stacking, and higher crystallinity. This demonstrates that bend point design serves as a core strategy in backbone engineering, precisely disrupting coplanarity through regulation of non-covalent interactions (spatial

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repulsion, S···O bonds). O locks) to precisely disrupt coplanarity, providing a molecular design rationale for the controlled introduction of bending points in flexible.

2.2. Dynamic key regulation strategy

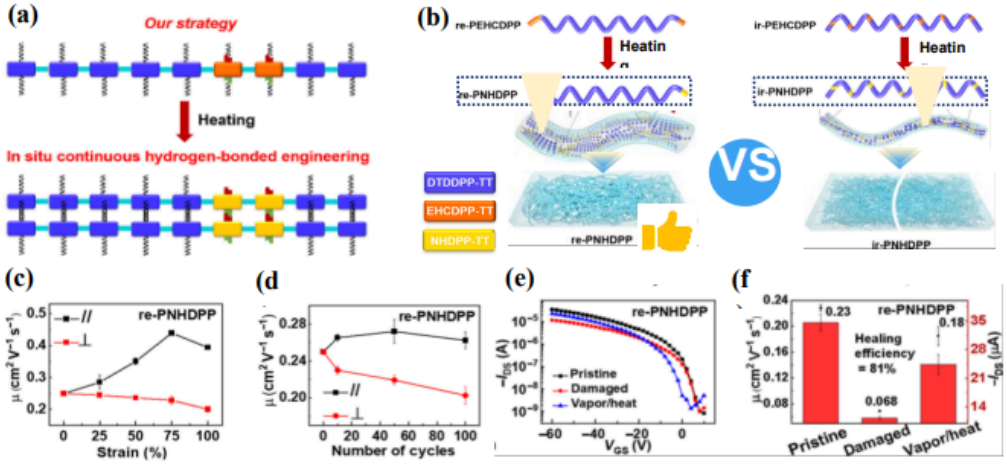

Figure 2. In situ continuous hydrogen-bonded engineering for intrinsically stretchable and healable high-mobility polymer semiconductors. (a) Schematic of the method. (b) Microscopic comparison of re-PNHDPP and ir-PNHDPP. (c) Mobility under stress. (d) Mobility versus stretching cycles. (e) Transfer curves of pristine, damaged, and repaired films. (f) Field-effect mobility and current of pristine, damaged, and repaired films [9]

The introduction of dynamic reversible interactions into the main chain or side chains, which dissipate energy through a break-recombination mechanism, can confer ductility or self-healing properties upon materials. Such interactions include hydrogen bonds, metal-ligand coordination bonds, and π-π bonds. Cao et al. enhanced the thiophene-pyrazine dihedral angle rotational energy barrier by 0.25 eV through intramolecular hydrogen bonding within the main chain thiophene-pyrazine units [8]. Results demonstrated that hydrogen bonding maintained device mobility without decay prior to 430K. Molecular dynamics simulations confirmed hydrogen bonds narrowed the main-chain dihedral angle distribution, preserving electron delocalization while preventing excessive stiffening [8]. This study pioneered defining main-chain torsion as a third phase transition independent of glass transition, expanding the theoretical scope of kinetic regulation. Additionally, employing reversible non-covalent bonds to enhance polymer toughness constitutes another strategy. When subjected to external forces, these relatively weak reversible non-covalent bonds fracture preferentially over the strong covalent bonds in the polymer backbone. This process dissipates the energy generated by the external force, mitigating stress concentration in the main chain. Yue et al. incorporated structural units capable of forming continuous hydrogen bonds into the polymer backbone. Employing a stepwise polymerization followed by thermal conversion, they generated a dense hydrogen-bonded network without disrupting the backbone conjugation [9]. As illustrated in Figure 2, re-PNHDPP exhibited an 1.8-fold enhancement in hole mobility at 75% strain—the highest improvement reported to date. Post-scratch heating enabled hydrogen bond reformation, restoring mobility to 81% of its initial value upon self-healing. This demonstrates that dynamic hydrogen bond engineering can confer both high elasticity and self-healing capability to polymers without sacrificing conjugation. However, this strategy has limitations. For instance, extensive

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introduction of dynamic non-covalent bonds may reduce material crystallinity and carrier mobility, as these weak bonds often accompany flexible groups that disrupt existing stacking order. Furthermore, the dynamic bonds may become ineffective under high temperatures or extreme conditions, necessitating comprehensive consideration of operating parameters.

2.3. Topological decoupling strategy

The core objective of topological decoupling is to eliminate mechanical coupling between D-A units through molecular structural design, thereby introducing stretchability while maintaining high mobility. To resolve the conflict between high mobility and high extensibility, one approach involves separating the stress-bearing flexible unit from the rigid functional domain that transmits charge. Alternatively, entropy/enthalpy regulation can be employed to disperse strain energy, with flexible linkers dissipating strain energy through chemical bond rotation. Although the synthesis process is relatively mature, the flexible unit disrupts conjugated continuity, resulting in mobilities generally below 0.1 $cm^2 V^{-1} s^{-1}$, making it difficult to withstand multi-directional strain. Rigid functional domains struggle to accommodate multi-directional strain scenarios due to high crystallinity. Alternating rigid functional domains with flexible linker bridges to form alternating rigid-flexible structures enables synergistic optimisation of electronic and mechanical properties at the molecular level. For instance, Lee et al. achieved sequential copolymerisation of rigid D18 and flexible PEHDT segments in OSCs. By independently synthesising D18 and PEHDT segments before block copolymerisation, they realised an alternating topological arrangement of rigid and flexible segments, thereby avoiding conjugation disruption and crystallinity reduction caused by random copolymerisation [10]. Results demonstrated that IS-OSCs retained 80% of their initial power conversion efficiency (PCE) under 31% strain, indicating that decoupling the topology of rigid and flexible segments can synergistically enhance both optoelectronic and mechanical properties. Additionally, Wu et al. proposed a strategy to achieve flexibility by modifying intrinsic material properties, without requiring external additives or macroscopic structural interventions [11]. Specifically, they induced molecular chain entanglement using an ultra-high molecular weight pure polymer (P4 with Mn = 97 kg mol^(−1)), enabling the rigid D-A backbone to dissipate strain energy through entropic elasticity, thereby achieving physical decoupling of the main chain [11]. As shown in Figure 3, with the rigid backbone intact, the charge transport pathways remain intact. Compared to conventional flexible bridging strategies, this approach endows the polymer with intrinsic stretchability (achieving 400% area expansion without fracture under 100% biaxial strain) while maintaining high charge mobility (mobility stabilised at 0.15–0.25 $cm^2 V^{-1} s^{-1}$). This offers a more streamlined material design pathway for high-performance stretchable electronics. However, for block copolymerisation approaches, achieving high molecular weight while ensuring controllable phase separation scales remains challenging. Otherwise, poor compatibility may compromise electrical performance. Conversely, for high molecular weight entanglement strategies, synthesising and processing ultra-long-chain polymers proves exceptionally difficult, with high-viscosity solutions hindering printable film formation. Moreover, such strategies often prioritise enhancing mechanical properties; should improvements to electrical properties prove limited, they must be combined with other approaches to achieve comprehensive results.

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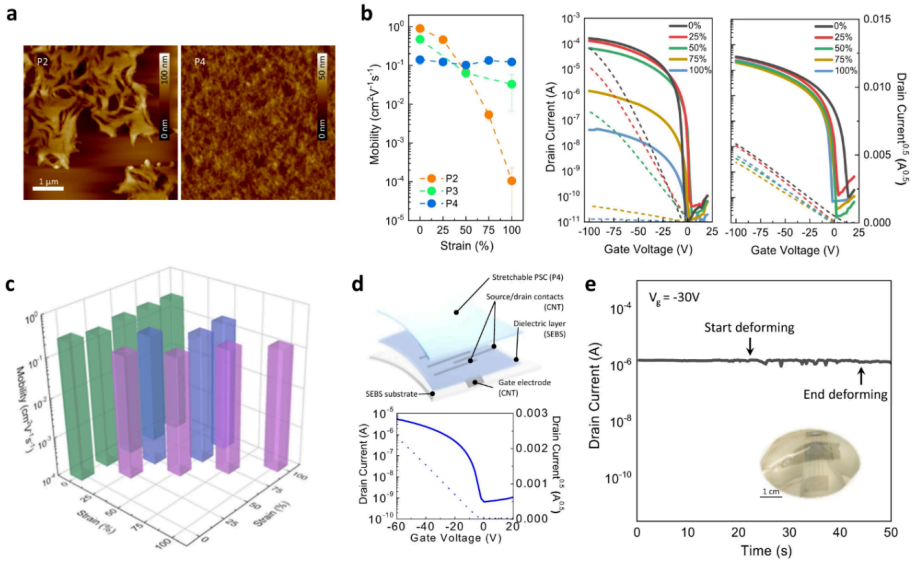

Figure 3. Highly stretchable polymer semiconductor thin films with multi-modal energy dissipation and high relative stretchability. (a) AFM images of P2 and P4 under 100% biaxial strain. (b) Average mobility and transfer curves of PSC FETs under biaxial strain. (c) Mobility of P4 under uniaxial vs. biaxial strain. (d) Architecture and electrical traits of stretchable P4 FETs. (e) Device stability under irregular strain [11]

3. Side-chain engineering molecular design

Compared to direct modification of the conjugated backbone, side-chain engineering offers a more diverse and gentle approach to enhancing the intrinsic flexibility and extensibility of D-A conjugated polymers by adjusting the structure and function of non-conjugated side chains. Side chains not only influence polymer solubility and processability but also significantly dictate interchain packing modes, crystalline domain size, and phase separation morphology, thereby indirectly regulating charge transport and mechanical behaviour in films. This section discusses three primary side-chain design strategies.

3.1. Regulation of flexible/dynamic side-chain units

Traditional conjugated polymers often incorporate long alkyl side chains to enhance solubility, yet these long, straight alkyl chains readily promote regular stacking of the main chain during stretching, leading to microcracks. Extensive research has developed multiple methods to regulate the flexibility of polymer semiconductors, with particular focus on controlling side-chain morphology through siloxane chains, silane chains, or embedded elastomeric segments. Siloxane side chains, owing to the high angular flexibility of siloxane bonds, are widely employed to enhance polymer mechanical flexibility and processability. For instance, introducing short oligomeric siloxane chains promotes separation between the main chain and side chains, reducing π-π packing distances and thereby increasing mobility. However, elongated or branched siloxane side chains exacerbate backbone distortion, leading to a significant decrease in mobility despite improved solubility. To balance these properties, Vantomme et al. explored combining siloxane with other flexible side chains, markedly enhancing polymer solubility [12]. As illustrated in Figure 4, introducing dynamic oDMS siloxane segments onto the DPP-TT conjugated backbone, coupled with long side chains and a branched structure, markedly reduces Young's modulus. This enhances the

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material's capacity to withstand greater deformation (branched oDMS (Si_7B) increases fracture strain to 14.5%) and confers intrinsic flexibility (the oDMS side chain exhibits a glass transition temperature below room temperature, Tg = −74 °C), thereby overcoming the rigidity limitations inherent in conventional alkyl chains. However, the introduction of flexible side chains or elastomeric segments often reduces the polymer's crystallinity and the upper limit of carrier Hall mobility. Therefore, how to quantitatively balance the impact of flexible.

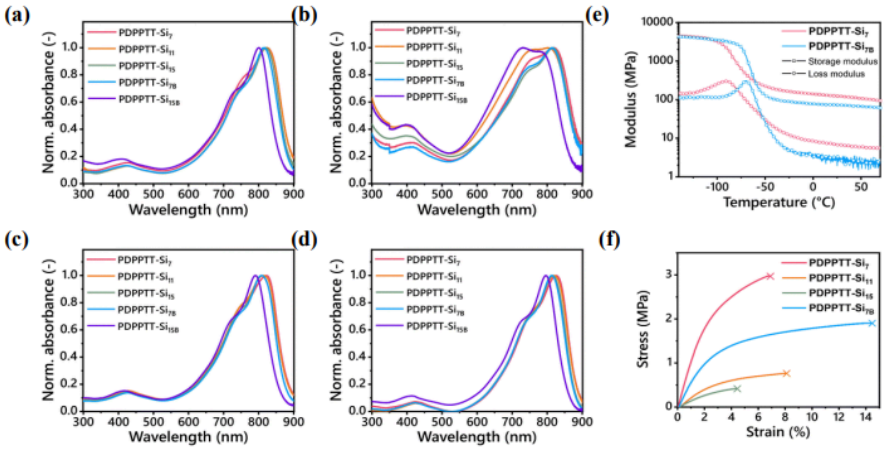

Figure 4. Trade-off between processability and device performance in donor–acceptor semiconductors revealed using discrete siloxane side chains. (a–d) UV–Vis spectra of PDPPTT-Si_n in four solvents (l = 1 mm, c = 0.08 mg mL^{-1}). (e) DMA curves of PDPPTT-Si7/Si7B (3°C min^{-1}, 1 Hz, 0.1% strain). (f) Stress–strain profiles of Si_n films from tensile tests [12]

3.2. Long branched side chains and crystallisation regulation

The primary objective of introducing long branched side chains is to enhance the polymer's solubility in organic solvents, thereby facilitating solution processing. Although the branching points generate significant steric hindrance that impedes the formation of compact, long-range ordered π-π packing structures along the main chain, leading to looser packing, reduced crystallinity, and improved film extensibility, the decrease in crystallinity weakens carrier transport capabilities. Thus, meticulous optimisation of side-chain length and density is required to achieve an appropriate equilibrium. For instance, Zhang et al. demonstrated through studies of side-chain engineered PDPPT polymers that branched structures offer advantages over purely flexible chains [13]. Research confirms that long branched side chains promote crystalline domain slip through lubrication rather than directly enhancing chain orientation, thereby achieving high extensibility (UV-vis spectroscopy indicates slightly higher orientation in long-chain polymers under high strain, while NEXAFS suggests minimal influence of side chain length on rotational behaviour) [13]. However, the quantitative relationship between branch chain length and aggregation behaviour, quasicrystalline states, and extensibility remains unclear. Consequently, Shih et al. systematically compared copolymers with varying branched alkyl chain lengths [14]. The study revealed that C_{20} long side chains mitigate tight aggregation, generating short-range ordered domains with substantial lattice disorder. This structure facilitates stress release during stretching, mitigating the disruption of long-range ordered regions [14]. As shown in Figure 5, the C_{20} sample maintains a high carrier mobility even at 60% strain, significantly outperforming the C_{12} short-chain polymer. PC20BTDFT (μ_h^// = 0.011 $cm^2\ V^{-1}\ s^{-1}$) exceeded PC12BTDFT (μ_h^// = 7.64×10^{-4} $cm^2\ V^{-1}\ s^{-1}$) by more than an order of magnitude [14]. This demonstrates that extending side-chain length can concurrently

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enhance both mobility and stretchability to a certain extent. However, longer side chains are not universally advantageous; excessive length may induce severe polymer chain amorphisation, significantly reducing mobility. Excessive size may also induce microscopic phase separation in the solid state, compromising film uniformity. Consequently, future research requires more detailed characterisation and simulation to elucidate how side-chain structural parameters influence interchain interactions, orientation changes, and crack initiation/propagation. This will guide the design of optimal side-chain lengths and branching configurations.

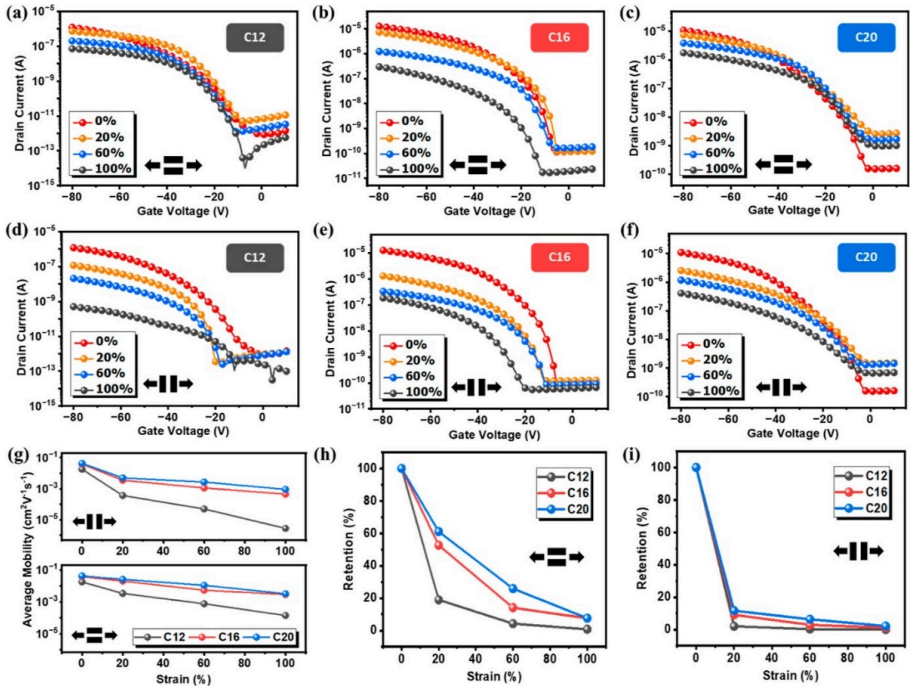

Figure 5. Revealing the effect of branched side chain length on polymer aggregation and paracrystallinity for improved mobility–stretchability properties. (a–f) Transfer characteristics of printed polymer FETs under 0–100% tensile strain. (g) Evolution of average hole mobility (μh) with strain, extracted from transfer curves. (h) Mobility retention of printed stretchable devices under tensile stress perpendicular to the stretching direction.(i) Mobility retention of printed stretchable devices under tensile stress applied parallel to the stretching direction [14]

3.3. Sidechain anchor positioning strategy

By regulating the connection sites and methods between side chains and the main chain, one can similarly exert a significant influence on the coplanarity of the polymer backbone, the degree of π-conjugation, intermolecular stacking, and electron cloud distribution, thereby controlling its optoelectronic and mechanical properties [15]. For instance, the electronic structural effects at side-chain anchoring points can be utilised to achieve a transition from a semiconductor to an intrinsically highly conductive state in polymers without the need for additional dopants. Ahn et al. synthesised two polymers based on the identical thiadiazole-benzoquinoline (D–A type) backbone: one featuring hydrophobic alkyl side chains on the quinoline acceptor units (PTQ-T), and another incorporating oxygen-affinity ethylene glycol side chains (PTQ-TEG) [16]. Despite sharing the same backbone, differing side-chain anchoring groups resulted in diametrically opposed properties, as illustrated in Figure 6. PTQ-T exhibits typical p-type organic semiconductor behaviour (with a hole mobility of 0.002 $cm^2/V \cdot s$ and an output curve displaying a distinct turn-off region).

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Conversely, the introduction of oxygen-rich ethylene glycol side chains confers PTQ-TEG with near-conductive high conductivity behaviour even without doping (the output current exhibits linear proportionality with voltage (VDS), with a conductivity of 3.0 × 10^{-4} S/cm, far exceeding conventional semiconductor CPs). This arises from oxygen atoms in the ethylene glycol side chains interacting with the main chain, enhancing the system's self-doped carrier concentration, increasing spin density, and inducing a transition from semiconductor to conductor. Concurrently, PTQ-TEG exhibits a smoother surface (the root-mean-square (RMS) roughness values for PTQ-T and PTQ-TEG are 0.478 nm and 0.355 nm respectively. Compared to PTQ-T, PTQ-TEG exhibits a slightly reduced RMS roughness), indicating enhanced molecular chain mobility, reduced rigid stacking, and favourable stress uniformity dispersion. This demonstrates the advantage of the side-chain anchor positioning strategy in balancing intrinsic flexibility with high mobility. Additionally, the conformation of the main chain can be controlled via the spatial positioning of the anchor site. Sabury et al. designed three isomers (PM7-D3, PM7-D4, PM7-D5) featuring ester side chains attached to distinct thiophene positions [17]. Results demonstrated that the side-chain anchor point position directly influences polymer backbone coplanarity and aggregation behaviour. PM7-D4 exhibited no aggregation in solution due to backbone distortion caused by its side-chain orientation, whereas PM7-D5's side-chain configuration favoured backbone straightness and strong aggregation, yielding the highest photovoltaic efficiency of 11.4% in devices. This reveals that the side-chain anchor point strategy can fine-tune polymer donor-acceptor compatibility and photovoltaic parameters. [17]. Concurrently, the substitution of rigid fused-ring structures with flexible thiophene bridges confers greater rotational freedom to the polymer backbone, thereby enhancing fracture strain resistance (the dihedral angle barrier θ_2 (thiophene-thiophene bond) for PM7-D5 is only ≈33°, significantly lower than that of conventional fused-ring polymers). However, current research on side-chain anchoring remains largely case-specific, lacking universal principles. For instance, further investigation is required into how to quantitatively predict the impact of a specific anchoring group on self-doping levels, as well as the coupling mechanisms between different anchoring positions and the main chain's conformation and extensibility.

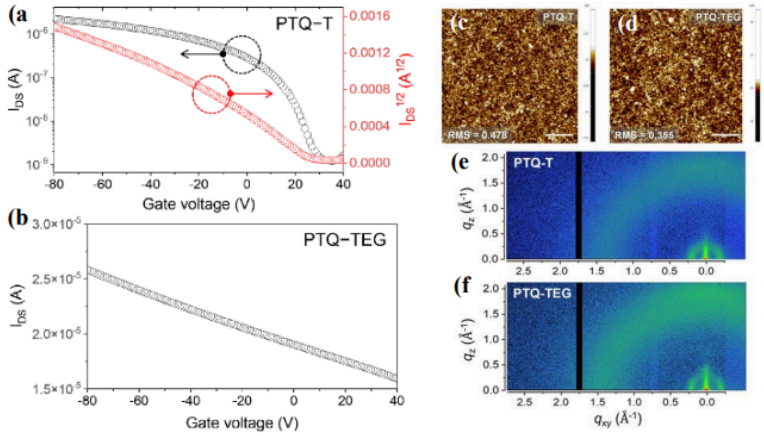

Figure 6. Side-chain-assisted transition of conjugated polymers from a semiconductor to conductor and comparison of their NO2 sensing characteristics. (a)–(b) Transfer characteristics of PTQ-T and PTQ-TEG FETs. (c)–(f) Surface morphology and molecular packing behavior of PTQ-T and PTQ-TEG characterized by AFM and 2D-GIXRD, respectively [16]

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4. Conclusion

In summary, this paper explores, from the perspective of molecular design, how synergistic innovation through backbone engineering and side-chain engineering achieves a balanced integration of optoelectronic performance and mechanical flexibility in D-A conjugated polymers. Regarding backbone engineering, the introduction of configurationally distorted units, the incorporation of dynamic bonds, and innovative topological structures have successfully mitigated the trade-off between high mobility and high extensibility to a certain extent. This enables materials to maintain excellent electrical properties while markedly improving mechanical flexibility. Regarding side-chain engineering, strategies such as flexible siloxane side chains, long branched side chains, and special anchoring groups provide richer regulatory means. These can significantly reduce polymer modulus, increase fracture strain, and in certain cases, achieve stable or even enhanced mobility with strain variation. Nevertheless, current research still faces challenges, including material trade-offs, complex synthetic methods, and the realisation of multifunctional synergistic optimisation. Future research must continue exploring more efficient molecular design strategies, particularly for enhancing the tensile properties of n-type conjugated polymers, while developing novel evaluation systems to better test and optimise the performance of stretchable optoelectronic devices. With ongoing advances in theoretical research and experimental techniques, D-A conjugated polymers are poised to play an increasingly significant role in next-generation flexible electronics, driving the development of wearable technology and smart electronic devices.

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