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REVIEW

Cyclopentadithiophene-based Conjugated Polymers for Organic Thermoelectric Devices and Other Applications

SeungOk Pyo[1], Yehbeen Im[2], Hyeokjun Kim[1*]

[1] Department of Organic and Nano Engineering, and Human-Tech Convergence Program, Hanyang University, 222 Wangsimni-ro, Seongdong-gu, Seoul, 04763, Republic of Korea
[2] Department of Chemistry, and Human-Tech Convergence Program, Hanyang University, 222 Wangsimni-ro, Seongdong-gu, Seoul, 04763, Republic of Korea

ABSTRACT

The 4H-cyclopenta[2,1-b:3,4-b']dithiophene (CPDT)-based conjugated polymers (CPs) have garnered significant attention in various fields of organic electronics due to their strong electron-donating properties, extended π-plane, and rigid, planar chemical structure. These unique features enable CPDT-based CPs to be highly advantageous for use in a range of organic semiconductor devices. While CPDT-based CPs have been extensively investigated and utilized as electron donors in various organic semiconductor devices, there is limited literature discussing the electrochemical properties of CPDT building blocks and the representative examples of CPDT-based CPs. In this mini-review, the authors outline the electrochemical properties of the CPDT building block, which stem from its rigid and planar chemical structure, facilitating the use of CPDT derivative materials in the field of organic semiconductors, such as organic photovoltaics (OPVs), organic thin film transistors (OTFTs), and organic photodetectors (OPDs). Furthermore, the authors highlight the advantages of CPDT-based CPs, particularly, for organic thermoelectric applications (OTEs) such as strong electron-donating properties and extended π-conjugation, which lead to facile p-type doping characteristics in CPDT-based CPs. The authors discuss the basic working principles of OTEs, including several key parameters of OTE devices such as the Seebeck coefficient (S) and power factor (PF). Additionally, the authors address the main challenge in OTEs: the trade-off relationship between electrical conductivity and the Seebeck coefficient. The review presents several strategies to overcome these trade-off limitations, focusing on CPDT and other CPs for OTE applications.

Keywords: Organic semiconductors; Conducting polymers; Doping; Quinoid; Seebeck effect; Power factor

*CORRESPONDING AUTHOR:
Hyeokjun Kim, Department of Organic and Nano Engineering, and Human-Tech Convergence Program, Hanyang University, 222 Wangsimni-ro, Seongdong-gu, Seoul, 04763, Republic of Korea; Email: gurwns6887@hanyang.ac.kr

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1. Introduction

For several decades, conjugated polymers (CPs) have garnered extensive interest for their potential as portable and flexible semiconductors in optoelectronic applications, including optical communications, energy storage, and light harvesting devices [1-9]. The appeal of CPs lies in their solution-processable nature, low toxicity, and ability to produce thin, lightweight devices. However, early CPs such as polyacetylene [10], poly(p-phenylenevinylene) [11], and polycarbazole [12] demonstrated relatively poor device performance due to inefficient charge transfer properties. To address this challenge, the D-A type copolymer, consisting of alternating electron-donating (D) and electron-accepting (A) moieties, has been rigorously explored. This structure promotes intermolecular charge transfer, extending π-conjugation in the polymer backbone through a push-pull effect, thereby enhancing charge transport properties in devices. Consequently, the selection of appropriate D and A moieties is crucial for optimizing CPs for optoelectronic applications [13]. Among various D moieties, the 4H-cyclopenta[2,1-b:3,4-b’]dithiophene (CPDT) moiety is one of widely used candidates, due to its extensive π-extended fused aromatic ring, strong electron-donating properties, low resonance energy of the aromatic ring, and facilitation of quinoidal population [14,15]. In practice, CPDT-based CPs have explored the applications with NIR absorption and narrow bandgap polymers. To achieve the NIR CPs and narrow bandgap polymers, the CPs should possess low bond length alternation energy [16], indicating an unstable resonance structure of CPs, leading to the ready formation of quinoidal structure. In this context, the inherent electron-donating property towards aromatic ring direction of CPDT moiety is highly advantageous for those applications. For instance, Yoo et al. reported a CPDT-based p-type CP that displayed NIR absorption from 700 nm to 1000 nm and ambipolar charge transport with high hole mobility of 1.50 $cm^2 V^{-1} s^{-1}$ [17]. Similarly, Bhat et al. developed an NIR organic photodiode (OPD) using a CPDT-based p-type CP, which exhibited broad absorption around 1200 nm and an NIR photocurrent responsivity of 0.4 $mA W^{-1}$ at –2 V under 1050 nm illumination [18].

Recently, CPDT building blocks have significantly contributed to the development of p-type conducting polymers (CPs) for organic thermoelectric (OTE) devices. OTEs are emerging as promising energy-harvesting devices for wearable electronics, converting human body heat into electrical energy. However, the inherent electrical conductivity of organic materials is typically too low for effective use in OTE devices. To overcome this limitation, the chemical doping method has been extensively researched [19-21]. This method enhances the electrical conductivity of organic materials by either losing (p-type) or gaining (n-type) electrons in organic semiconductors, thereby increasing the charge carrier concentration, which is directly proportional to electrical conductivity. In particular, the electrical conductivity of p-type polymer donors can be improved through reactions with strong oxidants such as $FeCl_3$ and F4TCNQ, which are recognized as p-type dopants. In the early stages of OTE research, benzene-based fused aromatic rings, including fluorene, benzodithiophene, and indacenodithiophene, were widely utilized as p-type building blocks for CPs. However, their limited effectiveness in p-type doping processes, due to the difficulty in extracting π electrons from their aromatized structures, resulted in poor electrical conductivity. Consequently, heterocyclic fused aromatic rings, such as CPDT-based building blocks, have been extensively employed. These rings have a relatively unstable aromatic structure that makes them more reactive towards p-type dopants. In this mini review, we introduce the chemical structure of the CPDT moiety and discuss its advantageous features for OTE devices. We explore the relationship between its chemical structure and electrochemical properties. Additionally, we introduce the fundamental working principles of OTE devices and highlight key parameters, including electrical conductivity, the Seebeck coefficient, and the power factor. Finally, we present examples of OTE devices that utilize CPDT-based CPs, along with their device performance.

2. CPDT-based CPs

In the early stages of developing conjugated polymers (CPs) and organic semiconductors, phenylene-based polymers like poly(paraphenylene) (PPP), poly(phenylene vinylene) (PPV) and poly(2,5-dialkoxy)paraphenylene vinylene (R-PPV) were explored as p-type donor polymers (**Figure 1**). These polymers were employed to improve the chemical stability of the polymeric chain and to enhance processability [22]. However, they exhibited limited charge transport due to their stable aromatized conjugated systems. To enhance the charge transport efficiency of CPs, five-membered heterocyclic compounds and their derivatives had been exploited, such as polypyrrole (PPy), polythiophene (PTh), and poly(3-alkylthiophene) (P3AT), because they have lower resonance energy compared to phenylene-based CPs due to its relatively high dipole moment toward aromatic ring structure (**Figure 1**). This unstable aromatized structure contributes to ready intramolecular charge transfer along the polymeric chains, leading to an efficient charge transport process. For instance, poly(3-hexylthiophene) (P3HT), a well-known p-type polymer, is extensively utilized in various organic semiconductor applications due to its facile processability, excellent crystallinity, and unique electrochemical properties. Nevertheless, with these unique characteristics of P3HT, the P3HT-based devices showed relatively low performance due to uniformly distributed charge carriers along polymeric backbone which limits intramolecular charge transfer, resulting in inefficient charge transport [23]. In this regard, the design strategy for efficient charge transport polymers has been extensively investigated. Among these demonstrated strategies, the structure of alternating electron-donating (D) and electron-accepting (A) building blocks, known as D-A polymers, has shown successful improvements in the performance of various devices. The D-A polymers possess numerous advantages for use as organic semiconductors, such as strong π-π stacking and vigorous intra- and intermolecular charge transfer, due to the dipole moment from the D to A building block, a phenomenon known as the "push-pull" effect. There are plenty of candidates for D and A building blocks, however, the thiophene-based heterocyclic building blocks have been investigated as D building blocks due to their strong electron-donating characteristics [24]. At the beginning of the development of D building blocks, thiophene and bithiophene were initially investigated due to their tendency to favor the elongation of π-conjugation. Despite these advantages of using thiophene and bithiophene as a D building block, the rotational freedom of each thiophene building block disturbs π-π stacking which leads to inefficient charge transport. For this reason, the fused thiophene derivatives, such as thieno[3,2-b]thiophene, have attracted significant attention from researchers to prevent their free-rotation. This strategy successfully elongates the π-conjugated length and extends π-planar to improve π-π stacking of CPs.

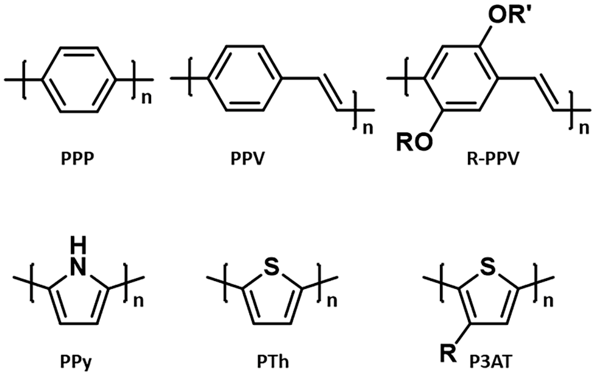

Figure 1. The chemical structure of phenylene-based (top) and heterocyclic-based (bottom) conjugated polymers.

Among those fused thiophene derivatives, CPDT is one of the representative thiophene-based fused aromatic rings. As illustrated in **Figure 2**, the π-extended conjugated structure allows the polymer to be rigid and planar, which contributes to be more ordered molecular structure and possesses better crystallinity, resulting in high electron mobility and charge transport properties in the devices. In addition, it has two acidic protons on methylene carbon which allow CPDT to be readily functionalized with soluble alkyl side chains. Moreover, because it has a relatively low resonance energy than benzene, it is advantageous to lose their π electrons through molecular doping process which is necessary to enhance the electrical conductivity of CPs [25].

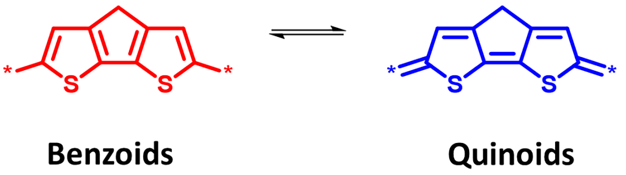

Figure 2. The chemical and resonance structure between benzoid and quinoid form of CPDT.

Since the first synthetic route of CPDT was reported by Kraak et al. in 1968 [26], and organic semiconductors have attracted interest in the past decades, CPDT has become one of the most widely used candidates for p-type organic semiconductors, and their electrical and optical properties have been investigated for utilization as electron-donating moieties in p-type polymers. Currently, a number of CPDT-based CPs have been developed for organic field effect transistors (OFETs), organic photovoltaics (OPVs), organic light emitting diodes (OLEDs) and OTEs. Lee et al. reported CPDT-based p-type polymers, **P1**, **P2** and **P3** via copolymerization between CPDT and diketopyrrolopyrrole (DPP) derivatives for OFETs (**Figure 3**). The solubility and crystallinity of **P1-3** were adjusted by the substitution of different types of alkyl side chain, resulting in different morphological and charge transport properties. The **P3** which consists of a combination between branch alkylated-CPDT and linear alkylated-DPP showed the most optimized morphology, highest hole mobility of 1.13 $cm^2 V^{-1} s^{-1}$, on/off ratio of 1.5×10^7, and V_{th} of 3.14 V [27]. Wang et al. reported CPDT-based polyelectrolytes, **P4** and **P5**, which were copolymerized with diphenyl ether or thioether, respectively (**Figure 3**). Those novel polyelectrolytes, which were doped by polyoxometalate, improved the performance of OPVs and OLEDs by increasing charge transport characters due to the high optical transparency, work function and electrical conductivity which dominantly contribute to extract and transport charge carrier, resulting in improved power conversion efficiency (PCE) of 15.3% with J_{sc} of 26.9 mA cm^{-2}, V_{oc} of 0.83 V and fill factor (FF) of 73% for **P5** based-OPVs and reduced turn-on voltage (V_{on}) of 2.8 V for **P5** based-OLED device [28]. Notably, various CPDT-based CPs have been developed to realize low bandgap near-infrared applications. Yoon et al. synthesized CPDT-based p-type low bandgap CPs, **P6** and **P7**, which are exploited for semitransparent organic solar cells (**Figure 3**). The synthesized polymers (**P6-7**) showed a strong NIR absorption peak around 760 nm in film state which indicates both polymers formed a low bandgap due to the extended π-conjugation and favor tendency to form quinoidal structure of CPDT. The fabricated semitransparent OPVs with **P7** showed PCE of 11.6% with J_{sc} of 24.06 mA cm^{-2}, V_{oc} of 0.70 V and FF of 69.1% [29]. The performance of the mentioned CPDT-based CPs is summarized in **Table 1**.

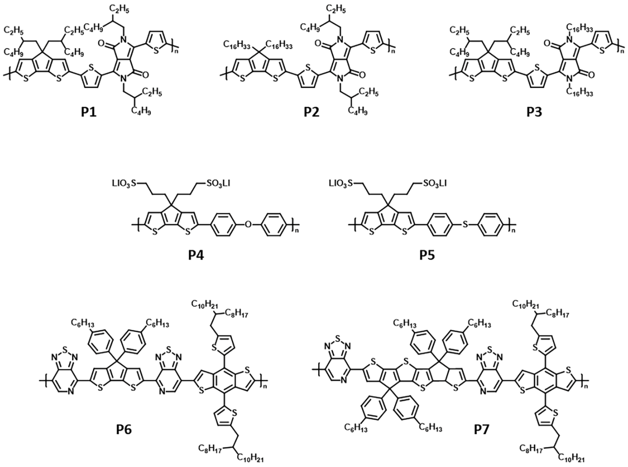

Figure 3. The chemical structures of CPDT based-CPs for various optoelectronic devices.

Despite the facile applications of CPDT-based CPs for various types of devices, they are more advantageous for use in OTEs due to their facile doping characteristics with p-type dopants. It is well-known that CPs can be oxidized or reduced by various doping methods, such as chemical and electrochemical doping. Among the various doping mechanisms in CPs, the integer charge transfer (ICT) doping mechanism is the simplest and most common process in OTEs. The ICT mechanism involves charge carrier exchange between CPs and dopants, resulting in the formation of polaron/bipolaron within the polymeric backbone of CPs and ionized dopant, as illustrated in **Figure 4**. To ensure that p-type CPs have higher electrical conductivity, it is essential to increase the charge carrier concentration along the polymeric backbone through the p-doping process. The strong electron-donating CPDT building block is advantageous for upshifting the HOMO levels of CPs, enabling CPDT-based CPs to be readily oxidized by p-type dopants, such as ferric chloride ($FeCl_3$) and F4TCNQ. In summary, CPDT-based CPs can be readily oxidized by various p-type dopants via the ICT doping mechanism, offering great potential for OTE applications.

Table 1. Summarization of introduced CPs and their device performances.

Device type	**Polymers**	**Hole mobility ($cm^2\ V^{-1}\ s^{-1}$)**	**On/off ratio**	**V_{th}(V)**			**Reference**
OTFT	P1	0.028	1.2×10^3	–4.78		**Annealed @150oC**	[27]
	P2	0.067	8.1×10^2	0.97			
	P3	0.94	4.5×10^3	1.98			
Device type	**Polymers**	**J_{SC} ($mA\ cm^{-2}$)**	**V_{OC} (V)**	**FF (%)**	**PCE (%)**		**Reference**
Perovskite Solar cell	P4	19.1	0.77	71	10.47	**Used as anode interfacial layer, Doped with POM**	[28]
	P5	22.8	1.14	78	20.1		
Device type	**Polymers**	**V_{on} (V)**	**Max L ($Cd\ m^{-2}$)**	**λ_{ems} (nm)**			**Reference**
OLED	P5	2.8	48,345	526		**Used as hole injection layer**	[28]
Device type	**Polymers**	**J_{SC} ($mA\ cm^{-2}$)**	**V_{OC}**	**FF (%)**	**PCE (%)**		**Reference**
OPV	P6	20.41	0.66	55.62	7.54	**Donor:Y6:PCBM ternary device , Transparent OPV**	[29]
	P7	24.06	0.70	69.12	11.62		

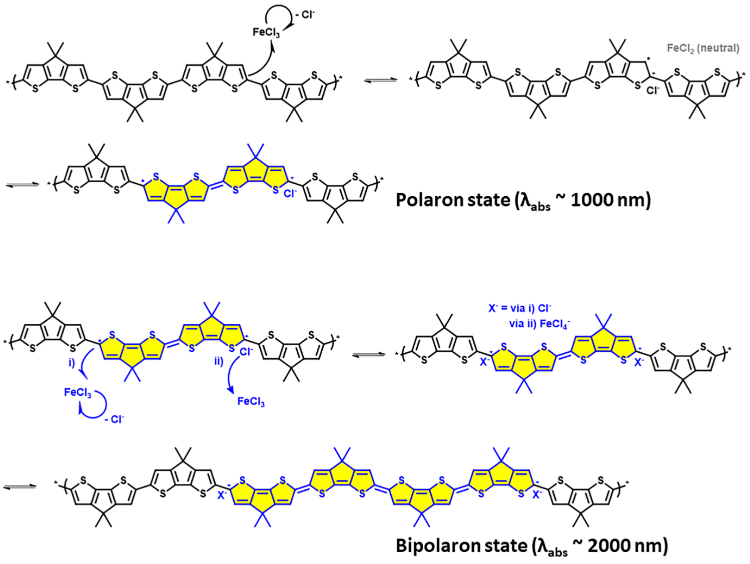

Figure 4. The schematic possible mechanism of forming polaron and bipolaron in conjugated polymer.

3. CPDT-based CPs for OTEs

With the rapid development of the electronics industry, electronic devices have become wireless, light, and convenient for use in wearable systems. However, current wearable applications rely solely on batteries as a power source, thus necessitating an additional continuous power supply. Notably, OTEs (organic thermoelectric devices) offer two advantages for use in wearable devices. First, OTEs possess mechanical softness, enabling electronic devices to be flexible and stretchable. Second, OTEs could theoretically provide permanent electricity to these devices by converting human body heat to power. The figure-of-merit, *ZT*, is a parameter used to estimate thermoelectric performance and can be expressed as Equation (1), where *S* is the Seebeck coefficient, *σ* is electrical conductivity, *T* is absolute temperature, and κ is thermal conductivity:

$$ZT = \frac{S^2 \sigma T}{\kappa} \tag{1}$$

Although *ZT* can quantitatively estimate the thermoelectric performance of metal or inorganic-based devices, organic-based thermoelectric devices are commonly evaluated by their power factor (*PF*), expressed as Equation (2), due to their intrinsically low thermal conductivity:

$$PF = S^2 \sigma \tag{2}$$

OTEs convert thermal energy to electric energy through the Seebeck effect, a phenomenon of converting a temperature gradient to electricity. As shown in **Figure 5**, one-component OTEs can be composed of a heat source, cold sink, and thermoelectric conducting polymers (CPs) with metal electrodes to measure *S* and *PF*. Once the temperature difference between the heat source and the cold sink is established and both ends are connected to doped CPs, the major charge carriers in the CPs travel through the polymer matrix with heat. In this case, charge carrier and heat conduction are dominated by κ and *σ*, respectively, expressed in Equations (3) and (4):

$$\kappa = \kappa_e + \kappa_L \tag{3}$$

$$\sigma = q n \mu \tag{4}$$

where κ_e is electronic thermal conductivity, κ_L is lattice thermal conductivity, *q* is the elementary charge, n is charge carrier concentration, and *μ* is carrier mobility. It is well known that κ_e is proportional to the *σ* of the material [30]. Hence, in the case of organic materials with intrinsically low *σ*, κ_e is negligible compared to κ_L, indicating that κ can be approximated to κ_L. This implies that thermal transport in the polymer matrix is predominantly determined by phonon transport. However, the mean free path of phonons in the polymer matrix is immensely short due to phonon scattering, crystalline defects, and grain boundaries in the polymer [31-33]. As a result, the thermal conductivity of CPs is extremely low compared with that of inorganic materials, often considered an advantage of organic materials for OTEs.

Figure 5. Schematic illustration of one-component OTE performance measuring system.

On the other hand, although *σ* can be improved by enhancing charge carrier concentration through chemical doping methods, high charge carrier concentration results in lower *S* due to their trade-off relationship [34]. For this reason, researchers have investigated new strategies for improving *σ* without a significant decrease in *S*. Cheon et al. synthesized a CPDT-based OTE CP, **T1** (**Figure 6**), which was doped with two different p-type dopants, $FeCl_3$ and tris(fluorophenyl)borane (BCF). The doped **T1** exhibited a highly ordered molecular packing structure even after doping, due to its local short-range ordered crystals in the amorphous region, regardless of the dopant type. As a result, the OTE performance of **T1** was measured as *σ* of 18.1 S cm^{-1}, *S* of 130 $\mu K\ V^{-1}$ and 30.7 $\mu W\ mK^2$ with $FeCl_3$ doping, and *σ* of 6.4 S cm^{-1}, *S* of 233 $\mu K\ V^{-1}$ and 35.1 $\mu W\ m^{-1}K^{-2}$ with BCF doping [21]. Another CPDT-based CP, **T2** and **T3**, were reported by Suh et al. (**Figure 6**). **T2** and **T3** were readily doped with F4TCNQ and exhibited enhanced morphological characteristics of PCT polymers by forming an ICT complex with the F4TCNQ anion. Notably, as dopant concentrations increased, the Seebeck coefficient was nearly maintained even with large amounts of dopant. Consequently, T3:F4TCNQ exhibited σ of 0.37 S cm^{-1}, *S* of 266 μK V^{-1}, and *PF* of 2.60 μW $m^{-1}K^{-2}$, and T3:$FeCl_3$ showed σ of 0.36 S cm^{-1}, *S* of 23 μK V^{-1}, and *PF* of 0.019 μW $m^{-1}K^{-2}$ [35]. As a p-type OTE example without a CPDT core, Qiang et al. synthesized two kinds of novel p-type OTE ladder-type CPs, **T4** and **T5**, which have different morphological characteristics (**Figure 6**). **T5** showed bimodal morphology containing both face-on and edge-on orientations, whereas **T4** exhibited only an edge-on orientation, indicating it was morphologically more uniform and relatively had fewer defects compared to **T5**. As a result, **T4** exhibited 7 times higher electron mobility than **T5**, and $FeCl_3$-doped **T4** achieved much higher thermoelectric performance, with σ of 36.29 S cm^{-1}, *S* of 315 μK V^{-1} and 22.7 μW mK^2, as the highest values [36]. As a challenging project, n-type OTEs have also been reported. Pei et al. reported the morphological properties of an n-type CP, **T6**, doped by N-DMBI (**Figure 6**). Although the **T6** pristine film exhibited a disordered molecular ordering pattern, the morphological character of T6:N-DMBI doped film was improved by enhancing molecular packing through intermolecular interactions between CPs and the dopant. As a result, the fabricated T6:N-DMBI-based OTE device achieved the best performance, with σ of 210 μK V^{-1} and *PF* of 25.5 μW mK^2 [37]. All the introduced OTE polymers above are summarized in **Table 2**.

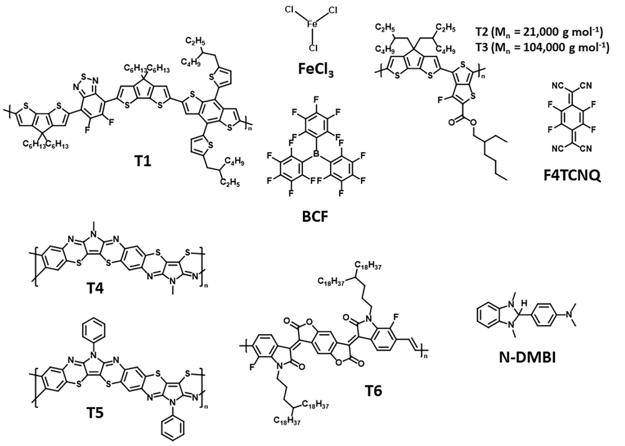

Figure 6. The chemical structures of organic thermoelectric polymers.

Table 2. Summarization of introduced CPs and OTE performances.

Polymers	**Carrier type**	**Electrical conductivity (S cm^{-2})**	**Seebeck coefficient (μ KV^{-1})**	**Power factor (μ Wm^{-1} K^{-2})**		**Reference**
T1:$FeCl_3$	**p-type**	18.1	130	30.7		[21]
T1:BCF		6.4	233	35.1		
T3:$FeCl_3$		0.37	266	2.60		[35]
T3:F4TCNQ		0.36	23	0.019		
T4:$FeCl_3$		36.29	315	22.7	**Varies on dopant immersion time.**	[36]
T5:$FeCl_3$		1.40	616	1.7		
T6:N-DMBI	**n-type**	6.23	–210	25.5		[37]

4. Conclusions

We have explored CPDT-based conjugated polymers (CPs), focusing on their structure, characteristics, and applications. The chemical structure of CPDT, featuring two lateral thiophenes and a central cyclopentadiene, forms a series of fused five-membered heterocyclic rings. This unique structure enables side chain modification via the two acidic protons on the central methylene carbon. As a result, CPDT-based CPs exhibit high oxidative potential and a quinoidal-favorable character, allowing CPDT an effective p-type building block for low bandgap CPs. In addition, we have reviewed the application of CPDT-based CPs in various optoelectronic devices, including OLEDs, OTFTs, and OPVs, with a special focus on OTEs. We provide a detailed overview of important parameters for OTEs, such as electrical conductivity, the Seebeck coefficient, and the power factor. While the power factor (*PF*) is directly related to electrical conductivity and the Seebeck coefficient, a trade-off between charge carrier concentration and the Seebeck coefficient often hampers OTE performance. Therefore, enhancing both electrical conductivity and the Seebeck coefficient in CPs requires careful doping and optimization of morphology in doped states.

Author Contributions

SeungOk Pyo mainly contribute to writing the original draft. SeungOk Pyo and Yehbeen Im reviewed and edited. Hyeokjun Kim is the corresponding author of this work and revised the final manuscript. All authors read and approved the final manuscript.

Conflict of Interest

The authors claimed no conflicts of interest.

Data Availability Statement

The data are available from the corresponding author upon reasonable request.

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