Communications in Computational Chemistry
Commun. Comput. Chem., Vol. 1, No. 2, 111–119
doi: 10.4208/cicc.2025.80.01
Regular Article

Unraveling the Impact of Non-Covalent Interactions and Different Donor Moieties on Charge Transport in DPPT-Based Polymers

Jinyang Luo[1], Jiawei Dong[2], Guo Wang[1], Linjun Wang[2,*] and Hua Geng[1,*]

[1]*Department of Chemistry, Capital Normal University, Beijing 100048, China;*

[2]*Zhejiang Key Laboratory of Excited-State Energy Conversion and Energy Storage, Department of Chemistry, Zhejiang University, Hangzhou 310058, China.*

[*] Corresponding authors: ljwang@zju.edu.cn, hgeng@cnu.edu.cn

Received on 21 March 2025; Accepted on 23 April 2025

Abstract: The integration of both rigid and flexible components holds great potential to significantly enhance the overall performance of organic electronic devices. Non-covalent interactions are frequently harnessed to augment the planar conjugation of polymers, consequently elevating the rigidity of these polymers. However, the influence of the dihedral angle distortion between donor and acceptor units, which is induced by the flexibility inherent in donor-acceptor copolymers, on charge transport remains poorly understood. In this study, we systematically investigate intra-chain charge transport parameters and charge mobility for the 3,6-bis(thiophen-2-yl) diketopyrrolopyrrole (DPPT) conjugated with various donor moieties. Combining with density functional theory (DFT) and the Su-Schrieffer-Heeger (SSH) model, we find that when the non-covalent interactions between the donor and acceptor units are enhanced (as exemplified by DPPT-FT and DPPT-BO), the coupling between electrons and low-frequency vibrations is significantly suppressed. Simultaneously, the intra-chain electronic coupling increases owing to substantial orbital overlap. Surface hopping simulations are utilized to study the charge transport properties. For DPPT-T, DPPT-FT, and DPPT-BT, weaker molecular rigidity and disordered chain packing lead to thermally activated hopping transport (low electronic coupling and high reorganization energy). In contrast, the enhanced structural rigidity of DPPT-BO facilitates charge delocalization, leading to an initial improvement in carrier mobility under low-temperature conditions, and thermal fluctuation effects induce a band-like behavior at high temperature.

Key words: Charge Transport, Diketopyrrolopyrrole Copolymers, Torsional Stiffness, Non-covalent Interactions, Su-Schrieffer-Heeger Model.

1. Introduction

Conjugated donor-acceptor (D-A) polymers as organic semiconductor materials have attracted much attention due to their promising applications in organic electronic devices [1,2]. The performance of these devices heavily depends on the charge transport properties of the D-A conjugated polymers. For these kind of polymers, covalent interactions establish main-chain backbone, determining its fundamental charge transport capability, while non-covalent interactions (including hydrogen bonds, van der Waals forces, π-π stacking, electrostatic