



Review

Recent advances in rylene diimide polymer acceptors for all-polymer solar cells

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ABSTRACT

In recent years, a large library of *n*-type polymers have been developed and widely used as acceptor materials to replace fullerene derivatives in polymer solar cells (PSCs), stimulating the rapid expansion of research on so-called all-polymer solar cells (aPSCs). In particular, rylene diimide-based *n*-type polymer acceptors have attracted broad research interest due to their high electron mobility, suitable energy levels, and strong light-harvesting ability in the visible region. Among various polymer acceptors, rylene diimide-based polymers presented best performances when served as the acceptor materials in aPSCs. Typically, a record power conversion efficiency (PCE) of 7.7% was very recently achieved from an aPSC with a rylene diimide polymer derivative as the acceptor component. In this review, we highlight recent progress of *n*-type polymers originated from two significant classes of rylene diimide units, namely naphthalene diimide (NDI) and perylene diimide (PDI), as well as their derivatives for aPSC applications.

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1. Introduction

Photovoltaic cells are attractive as a prospective technique to utilize solar energy, a renewable, clean, and non-fossil energy source. Widely investigated photovoltaic cells include dye-sensitized solar cells [1], quantum-dot solar cells [2], organic solar cells [3], and most recently developed perovskite solar cells [4]. Among them, organic photovoltaic (OPV) cells offering advantages of low cost, ease of processing, and flexibility over inorganic counterparts are most commonly fabricated with the photoactive layer comprising a blend of a conjugated organic semiconductor (OSC) and a fullerene derivative (bulk heterojunction (BHJ)) sandwiched between an anode (such as an ITO electrode) and a cathode (such as a metal electrode) [5]. The OSC serves as the donor material, in charge of the light absorbing and the hole transporting, while the fullerene as the acceptor material is responsible for electron transportation. When the donor material is a *p*-type semiconducting polymer, the polymer/fullerene blend-based devices are regarded as polymer solar cells (PSCs) [6].

In the PSCs, most studied fullerene-based acceptors such as PC₆₁BM and PC₇₁BM provide unique characteristics like favorable LUMO energy level, highly anisotropic electron transport, and good interactions with donor materials. In spite of these, some drawbacks of fullerenes for examples, invariable energy levels, limited light absorption in the red and near-infrared regions, and poor ther-

mal/photochemical stability, propel the search of their alternatives. Research efforts were thus devoted to employing *n*-type materials that could be either small molecules or polymers instead of fullerenes in the photoactive blend layer, developing the non-fullerene acceptors and corresponding OPV cells [7,8]. Compared to fullerenes and *n*-type small molecules, electron-carrying semiconducting polymers have unusual merits. First, the energy levels of conjugated polymers could be easily tuned via chemical modification. Secondly, such polymers can show broad absorption bands with high absorption coefficients covering the whole visible region by incorporating appropriate functional moieties into the polymers. Finally, donor polymer/acceptor polymer blends are beneficial for large-area solution processing thanks to the manageable solution viscosity [9]. Thereby, such BHJ OPV cells in which both donor and acceptor materials in the active layer are organic semiconducting polymers, namely, all-polymer solar cells (aPSCs), have been considered as a quite promising OPV technique for solar-to-electric conversion.

In the past few years, many *n*-type polymers have been synthesized as acceptor materials for aPSCs, based on electron-deficient building blocks such as cyano-substituted phenylenevinylene [10–15], benzothiadiazole [8,16,17], diketopyrrolopyrrole (DPP) [18,19], naphthalene diimide (NDI) [20], and perylene diimide (PDI) [21]. Among them, NDI- and PDI-containing polymers have been becoming the most promising candidates in terms of the fact that these two classes of *n*-type polymers are widely investigated and provide relatively excellent device performances for aPSCs [22,23]. In this review, recent advances in the NDI- and PDI-based and their derived polymer acceptors applied in aPSCs will be highlighted.

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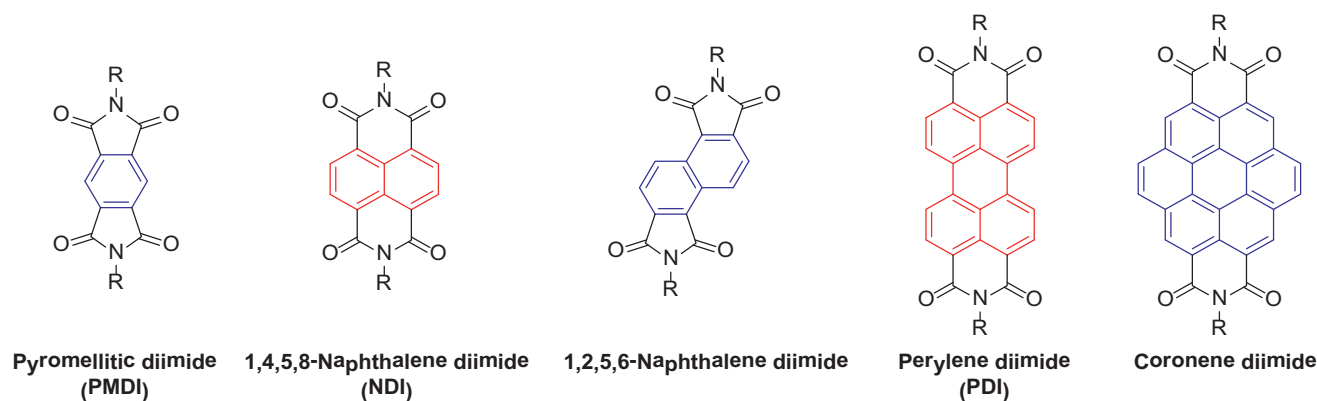


Fig. 1. Chemical structures of building blocks with electron-deficient tetracarboxylic acid diimide for *n*-type polymers. Among them, only NDI and PDI (with red cores) are strictly rylene diimide compounds, while others can be regarded as derivatives of the rylene diimide.

Operational principles of aPSCs are the same as those of typical BHJ OPV cells, which include four key steps: (1) generation of electron-hole pairs (excitons) via light absorption of the donor material; (2) diffusion of excitons to the donor-acceptor (D-A) surface and separation into free holes and electrons; (3) migration of free charge carriers to the contacts; and (4) collection of charges at the electrodes [7]. While each process must be optimized for improved device performance, other issues like excitons quenching and charge recombination have to be reduced as they will decrease the operation efficiency [9]. The cell performance is evaluated by the overall power conversion efficiency (PCE), the ratio of power output to power input, which is determined by three parameters, the short circuit current (I_{sc}), the open-circuit voltage (V_{oc}), and the fill factor (FF), achievable from the current (I) versus voltage (V) plot (I - V curve). The three significant parameters are closely related to the absorption behavior, HOMO/LUMO energy levels, and charge carrier transporting characteristics of the donor and acceptor species, as well as the blend morphology of the photoactive layer. Besides these, thermal annealing of the active layer, use of additives [24], introduction of different device structures such as inverted and tandem structures [25,26], and interfacial modification [27–30] also play crucial roles in determining the device performance. The detailed information about basic operation, performance parameters, optimization of device processing conditions, and device configurations can be found in previous publications [3,5,9,23]. On the other hand, synthetic details of polymer acceptors mentioned here, as well as device fabrication, were described in the original literature. In the following section, we focus on the molecular engineering that can efficiently adjust absorption feature and energy levels of the rylene diimide-containing *n*-type polymers (NDI- and PDI-based polymers and their derivatives), as well as on their performances as acceptor materials in aPSCs from a viewpoint of material chemists.

2. Rylene diimide polymer acceptors for aPSCs

Rylenes refer to hydrocarbon compounds which are naphthalene oligomers fused via 1,1' and 8,8' positions of adjacent naphthalene rings [23]. When two six-membered dicarboxylic imide rings are fused to the terminal naphthalenes, the rylene tetracarboxylic acid diimides (rylene diimides for short) are formed. The simplest rylene diimide, 1,4,5,8-naphthalene diimide (NDI in Fig. 1), together with its conjugation extended analogue perylene diimide (PDI) have been the most used building blocks for polymer acceptor materials. These two classes of polymer acceptors will be mainly emphasized in the present review.

Other three diimide compounds in Fig. 1 are not strictly the rylene diimide but can be regarded as their derivatives with similarly high electron affinity and strong light absorption due to the pres-

ence of the electron-deficient tetracarboxylic acid diimide units. Pyromellitic diimide (PMDI) has a smallest aromatic core, whose polymers should maintain the low-lying LUMO levels but suffer from challenging polymerization and backbone torsion due to the steric hindrance between the adjacent units. Although *n*-type PMDI-based polymers with alkyne units in the main chains, which could attenuate the hindrance through the polymer backbone, have been successfully prepared as field-effect transistor and thermoelectric materials [31,32], their uses as the acceptor species in aPSCs are to date not presented yet. 1,2,5,6-Naphthalene diimide, an isomer of 1,4,5,8-NDI, coronene diimide derived from PDI by fused two aromatic rings to the bay positions, as well as some other NDI-fused derivatives are also developed for polymer acceptors in aPSC. They will be discussed as well.

2.1. NDI-based polymers as acceptor materials

1,4,5,8-NDI has several advantages as a building block to make *n*-type polymers for aPSC. First, it is a highly coplanar electron-deficient aromatic core, which is a prerequisite to design *n*-type materials. Thereby, NDI polymers should be promising electron-carrying semiconductors when copolymerizing with appropriate comonomers [33]. Second, the imide position can be easily modified with various substituents for improved solubility that is significant for solution processable materials [34]. NDI polymers should have higher molecular weights than its PDI counterparts (see next section) due to its smaller conjugated core. Moreover, the N-substitution can adjust the polymer crystallinity, an especially important factor for morphological control and phase separation in the blend polymer film. Third, NDI polymers can be prepared with a regioisomerically pure form because of easy access to the 2,6-positions functionalized NDI monomer, which should also facilitate the formation of planar polymer backbone [22]. A variety of NDI polymers have been designed and synthesized, from which promising aPSC performances have been achieved.

A star NDI-based *n*-channel polymer is the P(NDI2OD-2T) (1, Fig. 2), also known as a commercial name N2200 [35]. It was reported with high electron mobility and has been widely investigated as the acceptor material in aPSCs. In an initial study, polymer 1 was blended with P3HT for the test of aPSC, yet giving a low PCE of only 0.16% [36]. Later on efforts were made to use different processing solvents and solvent mixtures for improving the efficiency of the 1/P3HT blend system [37], and a PCE of 1.4% was finally obtained using the mixed solvents of *p*-xylene and chloronaphthalene [38]. In recent studies, polymer acceptor 1 was paired with benzo[1,2-*b*:4,5-*b'*]dithiophene (BDT)-containing donor-acceptor polymers as the donor component in aPSCs. Based on the BDT polymers/1 blend system several high PCEs between 4% and 6% have been reported in the past 2 years

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