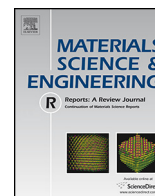




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# Organic polymeric and small molecular electron acceptors for organic solar cells

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### ABSTRACT

In organic solar cells (OSCs), the electron donor (D) and electron acceptor (A) blended active layer is the most crucial component for governing the power conversion efficiency (PCE). Various efficient donor materials with wide structural variations have been developed to couple with high-electron mobility fullerene-based acceptors, giving PCEs beyond 12%. However, fullerene-embedded OSCs encounter great challenges of low flexibility for structural modifications, poor absorption and blend morphological stability. The demand for alternative acceptors drives current OSC research towards non-fullerene acceptors (NFAs). Tailor-made NFAs of polymer or small molecule (SM) can typically exhibit tunable optical and electrochemical properties, high solubility, air stability, and favorable intermolecular interactions leading to compact packing and good nano-phase segregation in the active blend. In this review, we systematically depict the effects of molecular structures on the physical properties and device performances. The promising/most popular cores and general molecular design strategies of NFAs are outlined. The polymeric and SM NFAs were classified into several sub-groups based on their structural features, and in every sub-group, the structural evolution, current status, the champion case as well as the future challenges were highlighted and discussed. For polymeric NFAs, naphthalene diimide (NDI) and perylene diimide (PDI) are most promising and widely explored due to their easy synthesis, high electron affinity and mobility, leading to promising PCE when NDI and PDI units are conjugated with various electron-rich/deficient aromatics. Various electron-deficient core-based polymeric NFAs were also employed. Aromatic diimides (NDI and PDI) were also widely employed as the central core or terminal unit for SM NFAs. In particular, PDI was interested in electron deficient core, and their monomers, dimers, and trimers gave various degrees of success. PDI trimeric NFA showed superior PCE (~9.3%) because of its twisted 3D or fused geometry capable of interlocking the polymer donor allows optimum molecular packing, morphology and, therefore, efficient charge separation and transport. The excellent photochemical stability, strong absorption and synthetic flexibility of diketopyrrolopyrrole (DPP) produced promising SM NFAs. The rigid and co-planar indacenodithiophene (IDT) cores bearing various electron-deficient terminal groups were extensively explored, and the structural engineering on both the core and side chain groups together with post-treatments produced the highest PCE (~13.2%). These results conclude that NFAs possess the better possibility for tuning absorption profile, matched energy levels and optimal D/A nano-morphology for delivering promising PCEs. We highlighted the structure-property-performance relationships and future challenges, and hope this article can trigger new ideas for designing more promising NFAs.

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## 1. Introduction

Bulk heterojunction (BHJ) organic solar cells (OSCs) have attracted immense attention because they are potentially low-weight, cost-effective, semi-transparent, colorful, and flexible [1–3]. In OSCs, the electron donor and electron acceptor blended active layer is one of the most crucial component for governing the power conversion efficiency (PCE). Upon solar irradiation, the active layer absorbs sunlight, creates excitons (hole and electron pair weakly bound together) and diffuses at donor-acceptor interface into free hole and electron. Free charges are then transported, followed by collection at the oppositely charged anode and cathode, which generate the power by completing the electric circuit (Fig. 1). In 1986, Tang designed a bilayer OSC based on perylene tetracarboxylic derivative (PV) and copper phthalocyanine (CuPc) to achieve a PCE around 1% [4]. For the past two decades, various approaches have been developed for optimizing the active layer leading to a significant efficiency improvement [5–12]. Among them, the molecular design implementing various structural features onto respective constituents (donor or acceptor) generally plays the core role in this research field because the designated properties suitable for OSC applications can be feasibly and subtly tailored by chemical structures [10–12].

Several fullerene-based acceptors have been developed for OSCs, and their optimized devices showed high PCEs due to their several advantages, such as (i) high electron affinity, (ii) high electron mobility, (iii) electron transportation through its three-dimensional (3D) surface, (iv) multi-reversible reduction, and (v) ability to mix with donors and produce various mixed domains. Promising fullerene derivatives such as PC<sub>60</sub>BM, PC<sub>70</sub>BM and ICBA (Fig. 2) dominated other fullerene-based acceptors due to their high PCE till now [13]. However, they suffer from some significant drawbacks such as (i) high cost, (ii) limited absorption (UV region), which was the bottleneck for harvesting more photons from solar flux, (iii) difficult molecular designing towards red-shifted absorption, (iv) poor morphological stability due to aggregation tendency, which facilitates large size pure and mixed domains that

limit the exciton diffusion in an active blend. In the meantime, various efficient polymeric or small molecular (SM) donor materials with wide structural variations have been developed and coupled with high electron mobility fullerene-based acceptors to produce a PCE higher than 10% [11,12]. Nevertheless, OSCs employing conventional fullerene derivatives and the designated donor materials have reached the PCE plateau due to the difficulty in designing the molecules with spectral response covering the whole solar emission spectrum and simultaneously match to the energy levels of fullerenes. In addition, OSCs based on fullerene acceptors are generally suffering from the lower V<sub>OC</sub> as we expected from the energy difference between HOMO(D) and LUMO (A) due to poor interfacial charge-transfer states between the donor and the acceptor, high non-radiative recombination and non-geminate recombination [14,15]. To circumvent this limitation, OSC research is directed towards the development of new organic molecular acceptors, which are alternate to fullerene-based acceptors known as non-fullerene acceptors (NFAs).

NFAs potentially can retain the aforementioned advantages of fullerene-based acceptors along with additional benefits of (i) easy synthesis and purification process, (ii) tunable strong absorption with high molar extinction coefficients, (iii) efficient charge transfer with a low possibility of back electron transfer, (iv) higher solubility in common green solvents, (v) well mixing with donors and lead to compact packing and good nano-phase segregation in the active blend, and (vi) high morphological stability. In order to design the new NFAs, judicious selection of complementary donor was very important for a better spectral response of blend film covering the solar absorption region. Typically, donor and acceptor need a suitable energy levels alignment shown in Fig. 1b. The offset of highest occupied molecular orbital (HOMO) of donor and lowest unoccupied molecular orbital (LUMO) of acceptor directly governs the open circuit voltage (V<sub>OC</sub>) and, for effective charge transfer process ΔLUMO of donor and acceptor, should be in the range of 0.1 to 0.3 eV. Strategies to improve V<sub>OC</sub> can be made by raising the LUMO of acceptor and downshifting the donor HOMO with minimized

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