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Review

Non-fullerene small molecule electron acceptors for high-performance organic solar cells

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ABSTRACT

Fullerenes and their derivatives are important types of electron acceptor materials and play a vital role in organic solar cell devices. However, the fullerene acceptor material has some difficulties to overcome the intrinsic shortcomings, such as weak absorption in the visible range, difficulty in modification and high cost, which limit the performance of the device and the large-scale application of this type of acceptors. In recent years, non-fullerene electron acceptor material has attracted the attention of scientists due to the advantages of adjustable energy level, wide absorption, simple synthesis, low processing cost and good solubility. Researchers can use the rich chemical means to design and synthesize organic small molecules and their oligomers with specific aggregation morphology and excellent optoelectronic properties. Great advances in the field of synthesis, device engineering, and device physics of non-fullerene acceptors have been achieved in the last few years. At present, non-fullerene small molecules based photovoltaic devices achieve the highest efficiency more than 13% and the efficiency gap between fullerene-type and non-fullerene-type photovoltaic devices is gradually narrowing. In this review, we explore recent progress of non-fullerene small molecule electron acceptors that have been developed and led to high-efficiency photovoltaic devices and put forward the prospect of development in the future.

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1. Introduction

Since the disparity between economic development and environmental protection is becoming increasingly acute, people must look for a clean, renewable energy source to replace the fossil fuels. The research on clean and renewable energy sources has been one of the major technological challenges of the 21st century. Solar energy has aroused great attention of governments all over the world because of its non-geographical restrictions, no noise, no pollution, low cost and other advantages [1,2]. By 1954, the first silicon solar cell was fabricated at Bell Laboratories by Chapin et al. [3]. And its photoelectric conversion efficiency (PCE) reached 6%, which pioneered the study of photoelectric conversion. After that, the solar cell devices made of inorganic semiconductor materials have developed rapidly. In 1990s, GaAs, CdTe and laminated GaInP/GaAs/Ge devices were developed. They were composed of single crystals, polycrystalline or amorphous films. Although inorganic silicon solar cells have great potential for development, they are limited to such practical problems as complicated manufacturing process, high cost, high energy consumption and efficiency attenuation effect [4]. New generation solar cells include organic, inorganic-organic hybrid (e.g., inorganic-organic semiconductors), quantum dot, dye-sensitized, and perovskite (e.g., organic-lead halide hybrid) solar cells [5,6]. The development of coating and

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printing techniques make it economical and not trivial to manufacture organic photovoltaic cells (OPVs) by solution-processable materials [7,8]. The field of organic semiconductors is advancing rapidly, which promotes the popularization and application of low cost solar power generation technology. In stark contrast to traditional silicon based and other inorganic metal compounds solar cells, organic semiconductor solar cells have distinctive advantages, such as low energy consumption, wide range of sources and easy to implement large area and flexible devices [9–13]. Based on these advantages, organic solar cells have attracted people's attention and opened up a new direction of solar cells in the future [14].

Organic solar cells (OSCs) are devices which respond to light and convert light energy into electrical energy [15]. The device structure is typically made of indium tin oxide (ITO) transparent anode, metal cathode and photoactive layer sandwiched between them according to the composition of the active layer structure. OSCs typically employ p-n heterojunction device structures based on p-type donor materials and n-type acceptor materials. The whole process of the photoelectric conversion device can be divided into the following five steps: (1) optical absorption and exciton formation: when photons with energy beyond the absorption edge are incident on a semiconducting specimen of the polymer, an electron and hole with opposite spin are created, bound by their Coulomb attraction in a singlet exciton state and the absorption of sunlight in the bulk domains forms singlet excitons with different energy [16]; (2) exciton diffusion: after the generation of exciton, they diffuse through the donor (acceptor) phase and decay (radiatively or non-radiatively), or dissociate into free charges at donor/acceptor interface. Because excitons are neutral species, their motion is not influenced by an electric field and they diffuse passive; need to reach the heterojunction between the donor and acceptor components prior to their decay back to the ground state. This diffusion is typically described as a Förster-type incoherent energy transfer process, which can be either intramolecular or intermolecular and usually acts to lower the energy of the exciton. Because of the high exciton binding energy in conjugated polymers, the thermal energy at room temperature is not sufficient to dissociate a photo-generated exciton (typical binding energy of 0.4 eV) into free charge carriers. This downhill energy transfer can result in trapping of the exciton in the tail of the inhomogeneously broadened density of states. The trap sites are also often associated with defects and aggregates. At this point, any further exciton migration will rely on thermal fluctuations. At the same time, if the initially generated exciton is vibrationally excited, the diffusion process allows time for intramolecular vibrational relaxation to take place. Exciton dissociation can therefore occur from either the vibrationally excited "hot" exciton (i.e., the Franck-Condon state) or the thermally equilibrated, geometrically relaxed, "cold" exciton [17–19]. Hence, the thickness of the organic layers (or phases) has to be comparable to the exciton diffusion length L ($L = (D\tau)^{1/2}$, where D is the diffusion coefficient and τ is the lifetime of the exciton). A compromise regarding the thickness of the organic layers has to be found between allowing for efficient exciton diffusion to the heterojunction (favoring thin layers) and efficient sunlight absorption (favoring thick layers); (3) charge separation: after the generation of exciton, they diffuse through the donor (D) or acceptor (A) phase and decay (radiatively or non-radiatively), or dissociate into free charges at D/A interface. Upon reaching the D/A interface, excitons have to dissociate into electrons and holes. Initially, an exciton state at the D/A interface evolves into a charge-transfer (CT) state D^+/A^- , which then either recombines to the ground state or dissociates into free carriers via a manifold of charge-separated (CS) states [20]; (4) charge transport: once the charges have separated, they move toward their respective electrodes with an efficiency depending upon their mobilities.

In organic semiconductors, the electronic couplings (because of their intermolecular character), the large electron-vibration couplings (leading to marked geometry relaxations), and the disorder effects all conspire to produce more modest carrier mobilities because of charge carrier localization and formation of polarons. Charge transport then relies on polarons hopping from site to site. As a result, the charge-carrier mobilities strongly depend upon morphology and can vary over several orders of magnitude when going from highly disordered amorphous films (typically, $10^{-6} \sim 10^{-3} \text{ cm}^2/\text{V s}$) to highly ordered materials ($>1 \text{ cm}^2/\text{V s}$) [20]; (5) charge collection: electrons and holes reach the cathode and anode respectively to form an electric current. The efficiency of the charge collection process cannot be simply determined from the difference between the work function of the isolated electrode and the donor or acceptor [21]. An efficient collection of charges at their respective electrodes requires that the charge must overcome the potential barrier of organic/metal interface. Ion or metal diffusion into the organic layer, interfacial dipoles at the contact, band bending and chemical reaction can all affect the energetics at the inter-face and influence the injection process [22]. The total efficiency is the product of the five sub-process efficiencies [23–25], as shown in Fig. 1.

P-n heterojunction includes bilayer heterojunction (Fig. 2a) and bulk heterojunction (BHJ) (Fig. 2b). In the traditional OSCs before the mid-1980s, the current was generated by the metal/organics contact or differences in the work function of electrodes. Therefore, its photovoltaic properties depended strongly on the electrode, and the performance of these early organic solar cells was not satisfactory [26,27]. In 1986, Tang [28] introduced the concept of organic bilayer heterojunction for the first time. Bilayer OSCs were prepared and the PCE reached 1%, which had become a milestone in the research field of OSCs. However, bilayer heterojunctions are limited to vacuum deposited materials and the performance of this type of OSCs is limited by the exciton diffusion length (usually 5 ~ 30 nm) in the organic material [29]. Furthermore, the exciton diffusion length limits the maximum thickness of the active layer, which leads to low efficiencies due to weak light absorption.

In order to improve the PCE of OSCs, the most effective way is to expand the p-n interface to enhance the separation efficiency of electrons and holes. In a BHJ structure, the acceptor is completely doped in the donor material, both of which are fully mixed with each other to expand the p-n interface, more conducive to exciton separation. Excitons generated at any positions of the photosensitive layer in the film can all reach the interface of donor and acceptor material in a short way. This makes the separation efficiency between the charges to be largely enhanced, coupled with the positive and negative carriers formed at the junctions reaching the corresponding electrode position through a very short path, which fills up the deficiency of the carrier mobility [30,31]. Through spin-coating, printing and other means to process photosensitive active layer for OSCs, the BHJ structure provides the possibility for large area devices. Based on the above advantages, most of the efficient OSCs are now using this structure.

In 1992, Sariciftci et al. [32] found that the conjugated polymers and fullerene, [6,6]-phenyl-C₆₁-butyric acid methyl ester (PCBM, Fig. 3a) produced a rapid and efficient photo-induced electron transfer by photoexcitation. Since then, the fullerene typically attracted widespread attention and rapid development as an electron acceptor material. Subsequently, a series of fullerene derivatives had been developed, and exhibited good properties of electron acceptor. Hummelen et al. [33] changed the "carbon cage" structure by using C₇₀ instead of C₆₀ to prepare PC₇₁BM (Fig. 3b). The absorption of PC₇₁BM in the visible region was significantly stronger than that of PCBM and the PCE was significantly improved in PC₇₁BM based OSC devices [34]. However, due to the narrow range of absorption, the high affinity and the poor solubility of

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