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Review

Recent progress towards fluorinated copolymers for efficient photovoltaic applications

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ABSTRACT

This review paper summarizes the recent progress of highly efficient copolymers with the fluorination strategy for photovoltaic applications. We first present a brief introduction of the fundamental principles of polymer solar cells, and then the functions of fluorine atoms on the polymer donor materials. Finally, we review the research progress of the reported copolymers by classification of the fluorinated acceptor units and donor units, respectively. The resulting structure–property correlations of these copolymers are also discussed which shall certainly facilitate widespread utilization of this strategy for constructing high-performance photovoltaic copolymers in the future.

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1. Introduction

In order to resolve the problems of the traditional energy crisis and the consequent environmental pollution, efficient usage of solar energy by converting it into electricity is one of the most charming approaches. In the past few decades, great progress has been made in the field of photovoltaic applications. Recently, polymer solar cells (PSCs) have attracted the wide attention due to their advantages of low cost, flexibility, and light weight [1], which are regarded as the new generation of large-area solar cells by using the spin-coating, inkjet-printing, spray-coating, gravure-coating, or roller-casting processes, etc. [2]. At present, the bulk heterojunction (BHJ) device structure composed of both conjugated polymer donors and fullerene derivatives, where efficient charge separation is enabled by a large-area donor–acceptor (D–A) interface, has been regarded as the popular and effective architecture [3]. Enormous efforts have been made for integrative innovations on delicate material design as well as diverse device engineering to improve the photovoltaic performance of PSCs. State-of-the-art devices for single-junction PSCs have been certified records of over 10%, which is generally deemed as the entry point for commercial applications, although efficiency losses are still accounted from lab-scale cells to large-area modules [4–7].

1.1. Fundamental principles of PSCs

Typical PSCs possess a sandwich structure, where a photoactive layer is clipped between the cathode and anode. A relatively more complicated or efficient structure (Fig. 1) involves an additional anode or cathode interfacial layer (AIL/CIL) inserted between the photoactive layer and the electrodes [3,8]. Nowadays, the most studied PSCs are based on a BHJ configuration, where the photoactive layer contains an electron donor (D) and an electron acceptor (A). Four fundamental steps are included to describe the commonly accepted working mechanism of BHJ PSCs: (1) light absorption and generation of highly localized, tightly bounded Frenkel excitons; (2) exciton diffusion to the D–A interface; (3) exciton dissociation at the D–A interface: first creating charge transfer (CT) states or so-called polaron pairs, and then CT states fully dissociate into free charge carriers and (4) charge transport and collection [9–12]. Fig. 1 shows a schematic representation of above two typical BHJ device structures, illustrating the components involved in the mechanistic steps. The current–voltage curves also define the primary quantity and device performance of a solar cell [10,12]. A complete understanding of the working mechanism paves the way to realize the highly efficient PSCs.

In searching for high-performance PSCs, material designation attracted the continuous interests, and a great deal of research efforts in the last decades have been devoted to developing new conjugated polymer donors to extend absorption and harvest more solar energy. For an ideal polymer donor, appropriate energy

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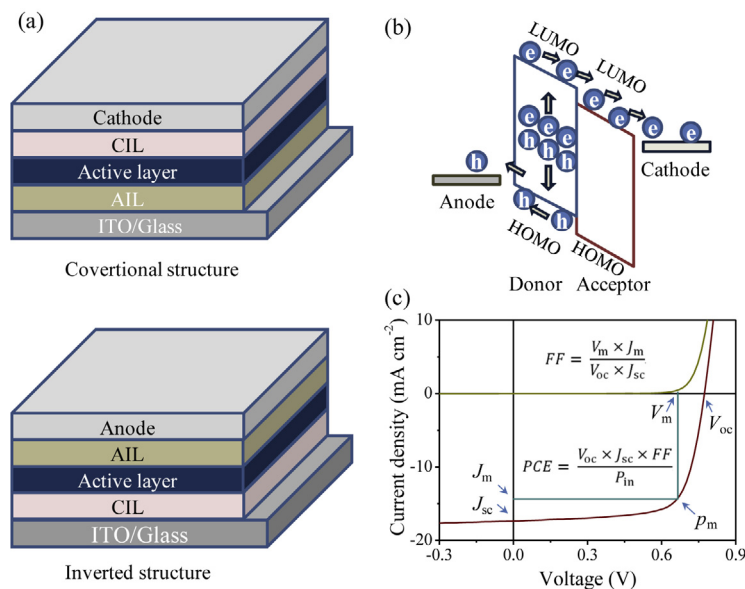


Fig. 1. (a) Device structures of PSCs. (b) The energy levels of the photoinduced charge transfer within PSCs. (c) J - V cures of a photovoltaic cell in the dark and under illumination.

levels, broad absorption ability and high charge carrier mobility are of great importance, which will directly affect open-circuit voltage (V_{oc}), short-circuit current density (J_{sc}) and fill factor (FF) of the related solar cells [13]. A deep HOMO level is desirable for obtaining a high V_{oc} because the V_{oc} of BHJ devices is directly proportional to the gap between the HOMO level of donor and the LUMO level of acceptor in the active layer [14,15]. In order to realize a satisfactory performance, polymer donors which contain an electron-donating (D) block and an electron-accepting (A) block have been tailored to improve the V_{oc} , while keeping a low band gap with broad absorption and thus allowing for a high J_{sc} . Rational choice of the D and A units to obtain a desirable “push-pull” effect of the alternating donor and the acceptor segments will induce an intra and/or intermolecular charge transfer (ICT), giving rise to a lower energy absorption band and a better photo-response [16]. In addition, the delicate side chain engineering would further finely tune the energy levels, crystallinity and solubility of the corresponding polymers [14,17]. Bear all above in mind, it is important to carry out detailed research work to explore the structure–property relationship of the photovoltaic polymers. Thus, a fundamental understanding of molecular designation and the benefits of versatile polymer syntheses allows for the effective tailoring of the intrinsic properties of photovoltaic polymers to serve for the desired purpose and address the application requirements [11].

1.2. The functions of fluorine atom in polymeric donor materials

Among the various material design strategies, introducing the fluorine atoms to the conjugated polymers has become a promising method for enhancing the efficiency of their polymer solar cells (PSCs). Fluorine atom has attracted wide attention as a modified group used for constructing highly efficient photovoltaic polymers due to many versatile unique characteristics: (1) fluorine atom is the smallest electron-withdrawing group with slightly larger van der Waals radius than that of hydrogen (1.35 Å vs 1.2 Å), and smaller than cyano and trifluoromethyl groups commonly used as electron-deficient substituents; (2) fluorine atom is the strongest electronegative element with a Pauling electronegativity of 4.0, much larger than that of hydrogen (2.2) [18–20]. It is well known that the introduction of an electron-withdrawing group into the polymer backbone can lower its energy levels [21]. Thus, the

fluorination can simultaneously lower the LUMO and HOMO levels of the resulting polymers while incurring only a minor effect on their optical band-gaps. As expected, BHJ PSCs based on these fluorinated copolymers would exhibit the higher V_{oc} and PCE values than the corresponding non-fluorinated derivatives [22,23]. Since it is only one small atom in size, it can be introduced onto the polymer backbone without any deleterious steric effects [24]. Moreover, an enhanced inter-/intramolecular interaction among polymer backbones can be found frequently due to the supramolecular interactions, such as C–F···H, F···S, C–F··· π_F , and so on [18,25]. The torsional angles within polymer backbones also would be minimized and the planarity should be enhanced [26]. Therefore, fluorination is always helpful to reduce intramolecular distances in comparison to non-fluorinated polymers, facilitating the formation of highly ordered solid-state structure with a more closed co-facial π - π stacking, leading to increase the crystallinity [20,27–29]. At the same time, these would lead to a more delocalized LUMO wave function that both polaron and exciton delocalization and their transport characteristics can be improved, resulting in higher carrier mobility for polymers and preferable morphology of their blend films with PCBM in PSCs, giving rise to the higher J_{sc} , FF and PCE [22,25,30–33]. In addition, fluorinated polymers exhibit great thermal stability and elevated resistance to oxidative degradation because the fluorine atoms can lower both the energy levels [18,29,34]. These features hold exceptional promise in enhancing the efficiency as well as the lifetime for polymer–fullerene PSCs, which are highly desired for practical applications [35].

In this review, recent progress towards highly efficient PSCs by rational design and reasonable modification of polymer donor materials with fluorination strategy on acceptor or donor skeletons are critically discussed, which will help us make deeply understanding of the fluorination effects and the structure–property relationships of the fluorinated photovoltaic polymers for efficient PSC applications.

2. Fluorination of acceptor building blocks

2.1. Thienothiophene-containing copolymers

Thieno[3,4-b]thiophene (TT) has been widely used for building D–A conjugated copolymers for PSCs. Low band-gap copolymers

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