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Review

Heavy metal complex containing organic/polymer materials for bulk-heterojunction photovoltaic devices

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ABSTRACT

The application of heavy-metal complexes in bulk-heterojunction (BHJ) solar cells is a promising new research field which has attracted increasing attention, due to their strong spin-orbit coupling for efficient singlet to triplet intersystem crossing. This review article focuses on recent advances of heavy metal complex containing organic and polymer materials as photovoltaic donors in BHJ solar cells. Platinum-acetylide containing oligomers and polymers have been firstly illustrated due to the good solubility, square planar structure, as well as the fairly strong Pt–Pt interaction. Then the cyclometalated Pt or Ir complex containing conjugated oligomers and polymers are presented in which the triplet organometallic compounds are embedded into the organic/polymer backbone either through cyclometalated main ligand or the auxiliary ligand. Pure triplet small molecular cyclometalated Ir complex are also briefly introduced. Besides the chemical modification, physical doping of cyclometalated heavy metal complexes as additives into the photovoltaic active layers is finally demonstrated.

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1. Introduction

Over the past few years, organic solar cells (OSCs) have attracted intense attention as renewable energy sources in both academia and industry, owing to their remarkable features such as flexibility, low cost, light weight, and large area mass production [1–5]. To date, the most efficient device structure for OSCs is utilized BHJ-type as the active layer [6–11], which consists of an interpenetrating blends of π -conjugated small molecules or polymers as electron donor (D) materials and typically fullerene derivatives [6,6]-phenyl-C₆₁-butyric acid methyl ester (PC₆₁BM) (Fig. 1) as electron acceptor (A) materials, providing efficient charge separation of the photogenerated excitons as well as favorable charge transport at the D/A interfaces [12–14]. The most common donor material in BHJ solar cells is regioregular poly(3-hexylthiophene) (P3HT) (as shown in Fig. 1), which can be used to achieve a power conversion efficiency (PCE) over 6% [15]. To acquire high performance BHJ solar cells, one of the most

successful methods is to modify the chemical structure of the donor materials to achieve the desired electronic properties. Although a large amount of new photovoltaic donor materials have been developed, small molecular OSCs with PCE over 9% have been rarely reported [16,17]; on the other hand, only a few polymers show PCE exceeding 10% in single-junction [18–20] and ~12% in multi-junction BHJ polymer solar cell devices [21–25]. In spite of the aforementioned success, after decades development of extraordinarily large number of organic/polymer donor materials, the possibility of further enhancements through chemical modification of designing new materials becomes a bottleneck [26,27]. Therefore, from the materials point of view, explore of new synthetic strategies affording extra approach to break through the PCE is particularly important. Since the mainstream of the previously reported organic/polymer materials all mainly based on singlet dominated pure organic materials, the introduction of organometallic heavy metal component with intersystem crossing is expected to better utilize the triplet excitons in devices to further improve the device performance. However, triplet concerning materials in organic photovoltaics (OPVs) have not been well investigated.

In general, the mechanism of a typical BHJ solar cell involves four steps: light absorption and exciton generation at the photoactive

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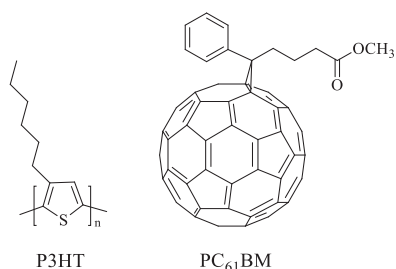


Fig. 1. Molecular structures of P3HT and PC₆₁BM.

materials, exciton diffusion to D/A interface, charge separation at D/A interface, and charge transport and collection [28]. Therefore, there are many aspects that can influence the performance of BHJ solar cells on the basis of the driving mechanism. Among, the average exciton diffusion length and charge separation are the most common factors to impact the power conversion efficiency. However, on one hand, the diffusion length (5–10 nm) [29,30] of singlet excitons in the most majority photovoltaic materials is usually significantly shorter than the absorption depth required for efficient light absorption. To increase the exciton diffusion length, the introduction of organic materials with high mobility and/or long lifetime is allowed [31,32]. On the other hand, in order to achieve high efficiency, eliminating geminate recombination of charge pairs that are formed by charge transfer (CT) at the donor-acceptor interface, which is competitive with charge separation should also be taken into consideration [33–36]. Besides, a major loss mechanism reported to limit the PCE of polymer solar cells is the “back reaction” in which the reverse energy transfer occurred from the photo-generated triplet charge transfer state (³CT) to the triplet state (¹T) of donor material when the triplet energy level values of CT state was lower than that of donor polymer [33,37]. Therefore, to reduce geminate charge recombination and facilitate charge separation, one possible way is to efficiently utilize the ³CT, as it is demonstrated that under strong magnetic field, the dark current of organic photovoltaic (OPV) devices could be increased by 45% due to the formation of triplet CT state [38]. In addition, ³CT with sufficiently long lifetime (micro-second scale or even longer) could increase the exciton diffusion lengths, making CT state more easier to separate and dissociate into free charges instead of recombination compared with the relatively shorter nano-second scale ¹CT state [31,33,39,40]. Thus, the formation of more ³CT based on heavy metal complexes is beneficial to increase the current and improve photovoltaic performance [31,39,40].

In the past decades, organometallic complexes have attracted extensive interests, owing to their specific interesting optoelectronic properties as the advanced characteristic to be widely applied as emitters in light-emitting devices [41,42], bioimaging [43,44] and sensitizers in dye-sensitized solar cells [45,46]. Whereas, organometallic complexes have been less investigated in organic thin film photovoltaics, and the initial studies have been focused on the donor/acceptor (*p/n* type) bilayer heterojunction device. For

example, Tang et al. fabricated the first organic photovoltaic cell with an conversion efficiency of 1% in a two-layer-heterojunction device by using phthalocyanine (CuPc) (Fig. 2) as the *p*-type active material [47]. In a recent development, Roy and co-workers reported the synthesis of tetramethyl-substituted Cu(II) phthalocyanine (CuMePc) (Fig. 2) nanocrystals and BHJ ternary solar cells based on CuMePc:P3HT:PCBM, displayed excellent photovoltaic performance with a conversion efficiency of 5.3% [48]. Besides, Shao and Yang demonstrated a simple multilayer heterojunction photovoltaic device with the configuration of ITO/PEDOT/PtOEP(30 nm)/C₆₀(30 nm)/BCP(8 nm)/Al(100 nm) [31]. The work based on typical triplet heavy metal complex 2,3,7,8,12,13,17,18-octaethyl-21*H*,23*H*-porphineplatinum(II) (PtOEP) as *p*-type donor material, achieved relatively high performance with 2.1% of power conversion efficiency. Furthermore, heavy metal based cyclometalated Ir complex mer-bis(4',6'-difluorophenylpyridinato-N,C₂') iridium(III) azaperylene (APIr) was also applied as the electron donor in a bilayer heterojunction device with a structure of ITO/APIr(5 nm)/C₆₀(30 nm)/N,N'-dihexyl-perylene-3,4,9,10-bis(dicarboximide) (PTCDI)(10 nm)/BCP(14 nm)/Al [49]. A fairly good PCE of 2.8% and considerably high open-circuit voltage (*V*_{oc}) up to 1.0 V was obtained.

In this review, recent progress on the development and application of heavy-metal complex containing photovoltaic donor materials for the bulk-heterojunction solar cells are presented. Low-bandgap platinum-acetylide [50–70] and the cyclometalated Pt or Ir containing polymers, oligomers or small molecules [71–77] as donor materials for the active layer of BHJ solar cells are successively demonstrated, in which the heavy metal complex was chemically bonded to the molecular backbone. Finally, physically doping of heavy-metal complexes as additives into the BHJ solar cells to enhance the photovoltaic performance are also discussed [78–81].

2. Heavy metal complex involved through chemical bonding

2.1. Platinum-acetylide containing polymers and oligomers

Platinum-acetylide containing derivatives is reported to be one of the effective *p*-type materials for organic solar cells, due to the square planar structure of Pt complex and fairly strong Pt–Pt interaction as well as excellent optoelectronic properties which is favorable to the performance of the photovoltaic devices [54,82]. The most promising and widely used platinum alkynyls have been a popular candidate for application into π -conjugated organometallic polymeric backbone [50–70]. The π -electron delocalization and the intra-chain charge transport along the entire polymer backbone is facilitated to be enhanced due to the *d*-orbitals (*d*_{xy} and *d*_{xz}) of the Pt atom overlaps with the π -orbitals (π_y^* and π_z^*) of the C≡C unit to construct the one dimensional conjugated polymer chain, when the alkyne is coordinated with the electron-rich Pt(II) ions [83]. Additionally, the presence of bulky PBu₃ ligands which prevents strong aggregation improves

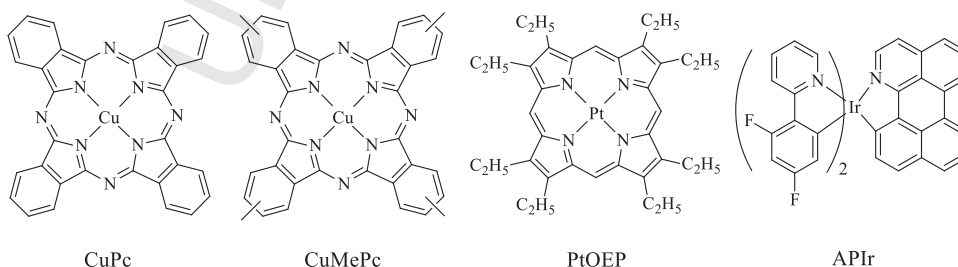


Fig. 2. Molecular structures of CuPc, CuMePc, PtOEP and APIr.

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