World Journal of Nanoscience and Nanotechnology

Review Article

Development History of Ternary Organic Solar Cells

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Abstract

Ternary organic solar cells (OSCs) have attracted the increasing interests as an effective method to improve photovoltaics performance. Moreover, ternary OSCs maintain sample fabrication technology as binary OSCs, which are of a great significance for the industry production. Here, firstly, we introduce the development history of ternary OSCs based on various systems. According to electron acceptors, ternary OSCs include two parts, that is involving only fullerene derivatives or at least one non-fullerene acceptor. Secondly, the Jsc evaluation is briefly described to further confirm the credible PCE of OSCs. Finally, we discuss some possible research directions of ternary OSCs in the future.

Introduction

In 1986, Tang [1] firstly reported the bi-layer organic solar cells with PCE of 1%, which is a milestone research in the field of Organic Photovoltaic (OPV). In 1995, Yu et al. [2] creatively fabricated a Bulk Hetero-Junction Organic Solar Cell (BHJ-OSCs) with PCE up to 3% by blending MEH-PPV and PCBM, which is a great breakthrough of OPV technology. From then on, BHJ-OSCs had attracted extensive attentions in the word due to many advantages of low cost, light weight, easy fabrication and mechanical flexibility [3-5]. The PCEs of BHJ-OSCs have exceeded 14% so far with the development of new materials, morphology engineering, interface modification and device architecture [6,7]. Insufficient photon harvesting of active layer is an important limitation for the PCE improvement owing to the intrinsically narrow absorption window of organic materials [8]. Ternary and tandem OSCs are designed to enhance photon harvesting by employing three or more materials with complementary absorption spectra. The middle connection layers are great challenge for highly efficient tandem OSCs, which need to low resistance, high light transmittance and ohmic contact with active layers. The complicated structures also add a lot of difficulty to obtain effective tandem OSCs [9,10]. Alternatively, ternary OSCs attract great interest of researchers by combining the advantages of tandem OSCs enhancing photon harvesting and single-junction OSCs with simple fabrication technology. Ternary OSCs are fabricated by incorporating the third component into effective binary active layers. Ternary OSCs can be divided in two categories: donor/acceptor$_1$/acceptor$_2$ (D:A$_1$:A$_2$) and donor$_1$/donor$_2$/acceptor (D$_1$:D$_2$:A) systems. According to the molecular weight, each system includes ternary polymer solar cells (PSCs) and ternary small molecule solar cells (SMSCs) [11,12]. The selection of the third component plays a vital role to obtain highly ternary OSCs, which need to consider absorption spectra, energy levels, compatibility of three materials. The versatile functions of the third component can be summarized as enhancing light absorption, optimizing film morphology, providing effective charge transport channels as well as improving device stability [13-15]. Many reports have demonstrated that the photon harvesting, exciton utilization, charge transport and collection in ternary active layer can be simultaneously improved by elaborately selecting the third component. Therefore, the photovoltaic parameters of short-circuit current density (J_{sc}); open voltage (V_{oc}) and fill factor (FF) can be synchronously increased for PCE improvement of ternary PSCs. To date, the PCEs of ternary OSCs have broken 14% by integrating the merits of three materials and two binary OSCs into one cell [16,17]. Ternary strategy is a simple and effective method to improve photovoltaic performance, which exhibits the bright perspective in the future industry production of OSCs.

In this contribution, firstly, we mainly introduced the development history of ternary OSCs. The universality and effectiveness of ternary strategy can be fully proved from ternary OSCs with various types. Secondly, the J_{sc} evaluation was briefly described to further confirm the credible PCE of OSCs. Finally, the summary and outlook of ternary OSCs were discussed.

Development History of Ternary OSCs

Ternary OSCs have become one of research hotspots in recent years with the growing number of publications, as shown in Figure 1a. Here, we summarized the development of ternary OSCs based on different types, as illustrated in Figure 1b.

Ternary OSCs involving fullerene acceptors

In 1995, Wudl et al. [18] invented one of the most important fullerene derivatives, PCBM. Since then fullerene derivatives ($PC_{61}BM$, $PC_{71}BM$, ICBA) have been the dominant electron acceptors of OSCs for about twenty years. The $PC_{71}BM$ is the most popular

Citation: Zhang M, Wang J, Qiaoshi An, Xiaoling Ma, Zhenghao Hu, Wang J, et al. Development History of Ternary Organic Solar Cells. World J Nanosci Nanotech. 2018; 1(1): 1002.

Publisher Name: MedText Publications LLC

Manuscript compiled: October 05th, 2018

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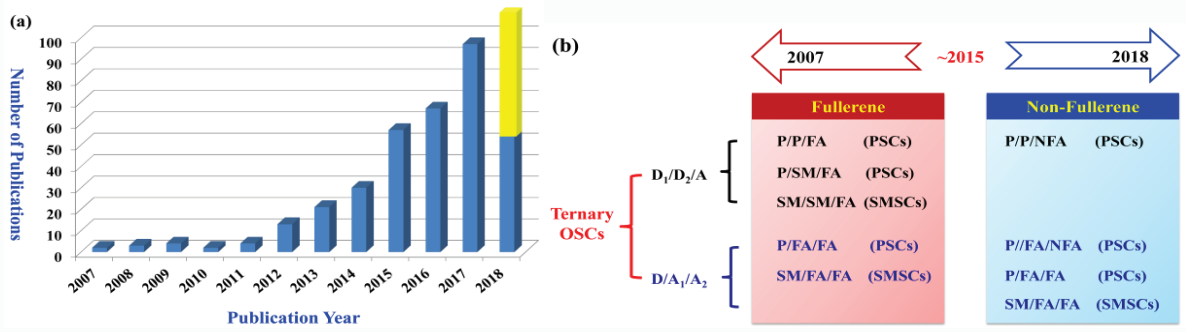

Figure 1: (a) The number of publications on ternary OSCs retrieved in Web of Science with "ternary solar cell" in tile as key terms; (yellow histogram represents the accepted articles in 2018 which can't be retrieved); (b) The classification of ternary OSCs.

acceptor for high-performance OSCs due to strong absorption from ultraviolet to visible light range and good morphology as blended with great majority of donors. The fullerene derivatives also have the shortcomings of high cost, difficulty synthesis and limited chemical modification. Therefore, a series of polymer or small molecule donors with different chemical and physical properties were developed to obtain highly efficient OSCs. Ternary OSCs especially based on D_1/D_2/A systems have been widely reported as an effective method to improve photovoltaic performance.

D_1/D_2/A based ternary PSCs (P/SM/FA and P/P/FA): The wide band gap polymer P3HT is the most commonly used electron donor materials [19-22]. At the early age, most of ternary PSCs are based on P3HT:$PC_{71}BM$ as the host system. The different types of donors were selected as the third component to improve the PCE of P3HT-based PSCs. In 2009, Honda et al. [23] reported dye-sensitized ternary PSCs with PCE of 2.7% by introducing the near-infrared dye molecule (SiPc) as the third component into P3HT/PCBM based host system. The SiPc molecules served not only as a photo sensitizer to enhance photon harvesting but as an energy transfer agent to improve P3HT excitons utilization. In 2010, Koppe et al. [24] firstly took a narrow band gap polymer PCPDTBT as the third component to improve the performance of P3HT/PCBM based PSCs. The charge transfer between PCPDTBT and P3HT can occur in ternary active layers, where hole and electron are mainly transported via P3HT and PCBM matrix, respectively. The PCE of ternary PSCs was improved from 2.5% to 2.8% with 20 wt% PCPDTBT content. In 2011, Xu et al. [25] synthesized insoluble CuMePc nanocrystals and incorporated it into P3HT/PCBM based PSCs to increase the carrier mobility and photon harvesting. The ternary PSCs with a P3HT:CuMePc:PCBM (1:1:2) composition exhibited the best PCE of 5.3%. In 2012, Yang et al. [26] firstly raised parallel-like bulk hetero junction (PBHJ) in TAZ/DTBT/PCBM and DTffBT/DTPyT/PCBM based two ternary PSCs. In this PBHJ, free charge carriers generated on two donors would travel through their respective corresponding donor/polymer networks to the electrodes, equivalent to a parallel-like connection. In 2013, Huang et al. [27] successfully constructed ternary PSCs with PCE 4.5% via incorporating squaraine dye SQ into P3HT/PC71BM based system. The energy transfer from P3HT to SQ was demonstrated by ultrafast TA experiments, which is an effective mechanism to enhance performance of ternary PSCs due to improved exciton migration over long distances. Later, narrow bad gap polymers have attracted great concerns due to their stronger light absorption in the Near Infrared (NIR) range and much deeper HOMO energy levels as well as better morphology with fullerene derivatives than P3HT. In 2014, Lu et al. [28] developed novel ternary PSCs by incorporating PID2 into a PTB7:$PC_{71}BM$ host system. The PCE of 8.22% was obtained for ternary PSCs with 10% PID2 in donors, mainly due to improved light harvesting, energy level cascading and optimized device morphology. In 2015, Zhang et al. [29] reported alloy-model ternary PSCs including one polymer (PTB7-Th), one small molecule (p-DTS-$(FBTTH_2)_2$) and one acceptor ($PC_{71}BM$). A notable PCE of 10.5% was obtained due to the enhanced crystallinity and the face-on orientation in ternary active layers by adding the high-crystalline small molecule of p-DTS-(FBTTH2)2. In 2016, Kumari et al. [30] investigated ternary PSCs comprising DR3TSBDT:PTB7-Th as a donor and $PC_{71}BM$ as an acceptor in detail. A state-of-the-art PCE as high as 12.1% was obtained from ternary OSCs with 25 wt% DR3TSBDT, which is the highest PCE value of fullerene-based ternary PSCs reported so far. In 2017, Zhang et al. [31] reported highly efficient and thick-film ternary PSCs (11.40%) by blending a nematic liquid crystalline small molecule BTR into PTB7-Th:$PC_{71}BM$ binary PSCs. The results show that the introduction of highly crystalline small molecule donors into ternary OSCs is an effective means to enhance the charge transport and thus is suitable to fabricate thick-films ternary OSCs for roll-to-roll production.

D_1/D_2/A based ternary SMSCs(SM/SM/FA): Compared with polymers, small molecules have many merits of high purity, well-defined molecular structure, definite molecular weight, less batch-to-batch variation and high charge carrier mobility [32,33]. The SMSCs have attracted more and more attentions while the PCEs of SMSCs are relatively behind of that of PSCs due to difficult morphology manipulation. The highly efficient ternary SMSCs are rarely reported. In 2015, An et al. [34] presented solution-processed ternary SMSCs with two low band gap small molecular materials (SMPV1 and DIB-SQ) as donors and PC71BM as acceptor. The PCE of 7.4% was obtained for ternary SMSCs by doping 10 wt% DIB-SQ in donors due to the synergistic enhancement of photon harvesting, exciton dissociation, charge carrier transport and collection. In 2016, Tan et al. [35] fabricated ternary SMSCs by combining DPPEZnP-THE(D_1) with $DPP(TBFu)_2$(D_2) and $PC_{61}BM$. The optimized ternary SMSCs produced over 7% PCE because of enhanced the crystallinity and domain purity in ternary active layer, which is rare in $PC_{61}BM$-based OSCs. In the same year, An et al. [36] achieved PCE of 10.16% for ternary SMSCs with DRCN5T and DR3TSBDT as donors, $PC_{71}BM$ as acceptor. In 2017, Nian et al. [37] reported highly efficient ternary SMSCs using two electron donors (p-$DTS(FBTTh_2)_2$ and ZnP)

and $PC_{71}BM$. Such a combination works well and simultaneously improved J_{sc} to17.99 mA cm^{-2} and FF to 77.2% of ternary SMSCs, yielding a milestone PCE of 10.97%. This PCE and FF is the highest value of ternary SMSCs reported so far.

$D/A_1/A_2$ based ternary PSCs and SMSCs (P/FA/FA and SM/FA/FA): Before 2016, the development of $D/A_1/A_2$ based ternary OSCs are limited owing to the lack of electron acceptors. The ICBA/$PC_{71}BM$ or ICBA/$PC_{61}BM$ is only possible selection to fabricate ternary OSCs. In 2011, Khlyabich et al. [38] reported P3HT/ICBA/PCBM based ternary PSCs and observed that V_{oc} of the ternary PSCs could be tuned between V_{oc} values of the corresponding binary PSCs. In 2013, Street et al. [39] proposed alloy model to explain the changed V_{oc} in P3HT-based ternary PSCs via charge transfer state measurement. In 2015, Kim et al. [40] chose two ternary SMSCs, DR3TBDTT:$PC_{71}BM$:$PC_{61}BM$ and DR3TBDTT:$PC_{71}BM$:ICBA, to investigate crystallization behavior and interactions among the three components. The nano phase crystalline structures were formed in two ternary active layers, resulting in the highest PCE of 3.04% and 5.11% of two ternary SMSCs, respectively. In 2015 and 2016, Cheng et al. [41,42] fabricated PTB7/ICBA/$PC_{71}BM$ and PTB7-Th/ICBA/$PC_{71}BM$ based ternary PSCs, respectively. The optimized ternary PSCs exhibit the PCE of 8.88% and 10.5%, which is mainly attributed to the alloy acceptor (ICBA/$PC_{71}BM$) formation for tunable V_{oc} and improved J_{sc} and FF.

Ternary OSCs involving non-fullerene acceptors

The non-fullerene acceptors (NFAs) have been studied since 1986 while the PCEs of NFA-based OSCs lag far behind that of FA-based OSCs for a long time due to poor active layer morphology [43,44]. Until 2015, Lin et al. [45] designed a star NFA, ITIC, which exhibits a PCE of 6.8% when blended with PTB7-Th, comparable to $PC_{61}BM$-based OSCs. The PCEs of NFA-based OSCs have increased dramatically since 2015 with effective NFAs similar to the ITIC structure [46]. A series of efficient NFAs further promote the development of ternary OSCs especially based on $D/A_1/A_2$ system.

$D_1/D_2/A$ based ternary non-fullerene PSCs (P/P/NFA): In 2016, Hwang et al. [47] reported the first highly efficient ternary non-fullerene PSCs with two polymer donors, PSEHTT and PBDTT-FTTE, and a new small molecule acceptor, DBFI-EDOT. The maximum PCE of 8.52% was obtained in the ternary PSCs with DBFI-EDOT:PSEHTT:PBDTT-FTTE=20:9:1 wt/wt, resulting from significantly increased J_{sc} due to extended the NIR absorption by PBDTT-FTTE. In 2017, Yu et al. [48] and Ma et al. [49] fabricated J51:PTB7-Th:ITIC and PTB7-Th:PBDB-T:IEICO-4F based ternary PSCs with PCE of 9.7% and 11.6%, respectively. In 2018, the PCE of 12.27% was obtained for ternary PSCs with PBDB-T:PTB7-Th:SFBRCN as active layers reported by Xu et al. [50], which is the highest PCE value of $D_1/D_2/A$ based ternary non-fullerene PSCs. Ternary all-PSCs have aroused wide concerns with the distinctive advantages of outstanding thermal and mechanical stabilities and facile modification of the chemical and electronic properties of active layer components. In 2016, Benten et al. [51] successfully fabricated ternary all-PSCs with PBDTTT-EFT/PCDTBT/N2200 as the active layers. The PCE of 6.65% was obtained for ternary all-PSCs containing 10 wt% PCDTBT in donors. In 2017, Li et al. [52] demonstrated that the PCE of PTB7-Th:PNDI-T10 all-PSCs can be improved by incorporating a polymer PBDTTS-FTAZ as the second donor. The optimized ternary all-PSCs attain PCEs of 9.0% in both the conventional and inverted devices. In 2018, ternary all-PSCs with a high PCE of 9.56% and an impressive FF of 75.7% were achieved by blending PBTA-Si with PTzBI-Si and N2200, which is the highest PCE and FF values of current ternary all-PSCs reported by Fan et al. [53] The NFAs are very picky with the donors, leading to the relatively large difficulty to obtain highly efficient $D_1/D_2/A$ based ternary non-fullerene PSCs.

$D/A_1/A_2$ based ternary non-fullerene PSCs (P/NFA/NFA): Various NFAs with different band gaps and energy levels provide enough chance to design P/NFA/NFA based ternary non-fullerene PSCs. Many NFAs exhibit good compatibility benefiting from their similar chemical structures, which is propitious to form good morphology of ternary active layers. In 2016, Baran et al. [54] reported the highly efficient and stable P3HT (7.7 ± 0.1%) based ternary PSCs in an inverted architecture combining with two non-fullerene acceptors of IDFBR and IDTBR. This discovery revives the use of P3HT in high-performance OSCs, reducing the efficiency-stability-cost gap of commercialized ternary PSCs. The PCE of ternary PSCs can arrive to 11.0 ± 0.4% by replacing the P3HT with the low-band gap polymer PCE10. In 2017, Jiang et al. [55] fabricated ternary PSCs with PBDB-T:ITM:ITCN as the active layers and a highest PCE of 12.16% was observed, which is higher than the PBDB-T:ITM-based PSCs with 10.89% PCE and PBDB-T:ITCN-based PSCs with 2.21% PCE. Zhang et al. [56] reported efficient ternary non-fullerene polymer solar cells with PCE of 11.92% and FF of 76.5%, which is attributed to the excellent compatibility of PBDB-T, IDT6CN-M and ITCPTC as well as complementary photovoltaic properties of two binary PSCs. In 2018, Ma et al. [57] reported PBDB-T:INPIC-4F:MeIC1 based highly efficient ternary PSCs by integrating the advantages of materials and two binary cells. The optimized ternary PSCs exhibit a high PCE of 13.73% with 50 wt% MeIC1 in acceptors, resulting from the simultaneously improved J_{sc} of 21.86 $mAcm^{-2}$, V_{oc} of 0.88 V and FF of 71.39%. Shortly afterwards, the PCE of ternary non-fullerene PSCs was further refreshed by Zhang et al. [58]. They employed a polymer donor (PBDT-T-2F) and two NFAs with similar chemical structure but very different photoelectric properties to fabricate ternary PSCs. The optimized ternary PSCs delivered a PCE of 14.18%, which is the top PCE value of ternary OSCs reported so far. The P/NFA/NFA based ternary non-fullerene PSCs might be the most promising candidates to achieve high PCE even over 15% for commercial standards.

$D/A_1/A_2$ based ternary PSCs and SMSCs (P/FA/NFA and SM/FA/NFA): Fullerene derivatives own the advantages of strong ultraviolet-light absorption, three-dimensional electron transport property and high electron mobility. NFAs have the advantages of strong absorption in visible and NIR region, easily tunable energy levels, controllable film morphology and improved stability. Combing fullerene derivatives with NFAs together to fabricate ternary PSCs might be a promising strategy to improve device performance. In 2016, Lu et al. [59] presented P/FA/NFA based ternary PSCs for the first time by blending $PC_{71}BM$ into PPBDTBT:ITIC host system. The PCE of ternary PSCs was largely boosted to 10.4% (≈ 35% enhancement relative to the control devices) by combining the merits of $PC_{71}BM$ and ITIC. Subsequently, Zhao et al. [60] improved the photovoltaic performance of PBDB-T:IT-M-based PSCs by introducing B is [70]PCBM as the third component. An outstanding PCE of 12.2% was achieved for the optimized ternary PSCs due to the enhanced photon harvesting and effective diffusion of exciton in ternary active layers. In 2017, Xiao et al. [61] designed an ultra-narrow band gap NFA of CO_i8DFIC, which exhibit a 10.48% PCE

when blended with the PTB7-Th. The PCE can be further increased to 14.08% by incorporating 30 wt% $PC_{71}BM$ in acceptors, which the highest PCE value of P/FA/NFA based ternary PSCs. In 2018, Gao et al. [62] also fabricated highly efficient ternary PSCs with PCE of 13.54% based on PTB7-Th:3TT-FIC:$PC_{71}BM$ as the active layers. The PCE enhancement was mainly ascribed to the improved EQE response in the visible range and charge transport in ternary active layers by blending the $PC_{71}BM$. Ternary PSCs combining fullerene derivatives with NFAs have been demonstrated as an effective strategy to improve device performance. In contrast, the SM/FA/NFA based ternary SMSCs are rarely reported owing to the absence of effective small molecule donors. In 2017, Zhang et al. [63] achieved ternary SMSCs with PCE of 10.48% consisting of two acceptors (IDIC and $PC_{71}BM$) and a donor (DRTB-T). The PCE of ternary SMSCs would be further improved in the future by synthesizing more effective small molecule donors matched well with NFAs.

The Evaluation of Short-Circuit Current Density

It is well known that the PCE of OSCs is determined by the product of J_{sc}, V_{oc} and FF divided by the input power (1,000 W/m^2). The J_{sc} is a relatively contentious parameter because many uncertain factors would lead to an inflated measured J_{sc} value, such as effective area, light intensity. It is very important to evaluate the J_{sc} accuracy for credible PCE of OSCs. Sending the prepared OSCs to a professional certification authority for PCE measurement might be the best method to demonstrate the real experimental data. So far, the method is difficult to put into effect due to the lacking of mature encapsulation technology and unified measurement standard in the organic photovoltaic field. Another feasible way is that using EQE spectra of OSCs to calculate the J_{sc}, which should be close to the measured J_{sc} of the corresponding OSCs with deviation less than 5%. The calculated J_{sc} of OSCs is equal to the integral of the product of EQE response and incident solar spectral irradiance at different wavelength. Figure 2a exhibits the calculated J_{sc}s of PSCs according to the AM 1.5 solar light radiation spectrum and assuming the EQE values of 75% in the whole spectral range. As seen from Figure 2b, the solid and dotted line represent the real and assumed EQE spectra of binary (black) and ternary PSCs (red), respectively. When the absorption edge of binary active layer is about 960 nm, the maximum theoretical calculated J_{sc} is 26.78 mA/cm^2. The maximum theoretical calculated J_{sc} can be slightly increased to 28.39 mA/cm^2 if the absorption edge is red-shifted to 1,000 nm by incorporating the third component. Obviously, the theoretical calculated J_{sc} should be much larger than the real value because the area of rectangle (EQE=75% and X axis) is much larger than the area surrounded by real EQE spectrum and X axis. The real J_{sc} values are increased from 22.98 mA/cm^2 of binary PSCs to 24.13 mA/cm^2 of ternary PSCs, which should be acceptable. If the measured J_{sc} is very close to theoretical calculated value, we should pay more attention to the measured J_{sc} to further check the measurement conditions.

Summary and Outlook

Ternary OSCs can improve device performance at the same time maintain simple fabrication technology, which is a great significant in future industry production. The future development of ternary OSCs may concentrate on the followings: (1) More functions of the third component should be explored to further improve the PCE of ternary OSCs by designing new materials; (2) The working mechanism of ternary PSCs need to be further investigated, including charge transfer, energy transfer, parallel-linkage structure, alloy model and coexist of multiple mechanisms; (3) The stability study of ternary OSCs should be carried out; (4) Ternary strategy could be used to fabricate highly efficient thick-film OSCs, large-scale OSCs or semi-transparency OSCs for diversified commercial applications of OSCs.

Acknowledgement

This research was funded by the Fundamental Research Funds for the Central Universities (2018YJS168), National Natural Science Foundation of China (61377029, 61564003, 61675017, 61705161).

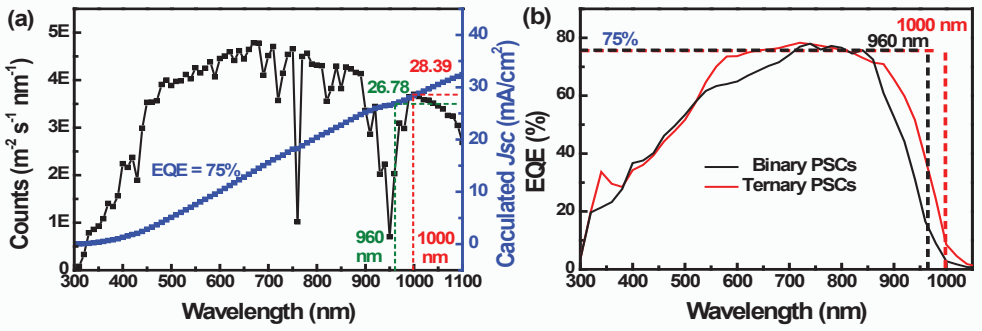

Figure 2: (a) The photon flux spectrum of solar light and theoretically calculated JSC; (b) The EQE spectra of binary and ternary PSCs.

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