The Optimization of Transparent Organic Photovoltaic Cells

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Abstract. As an emerging technology, transparent organic photovoltaic cell (TOPV) has the potential to diversify the applications of solar energy and extend its reach to additional fields. In the last dozen years, a multitude of innovative TOPVs have been crafted. Excellent TOPVs exhibit superior electrical conductivity and notable visible light transmittance, leading to increased power conversion efficiency (PCE) and average visible transmittance (AVT). In order to summarize the achievements of the existing technology of TOPV and provide directions for its future development, this review is authored. It revolves around three aspects: optimizing PCE and AVT separately and balancing high PCE and AVT concurrently. This review offers professional optimization strategies for the aforementioned three aspects and provides an overview of the most recent advancements in the field of TOPVs. Through the research results, PCE and AVT of TOPVs are developing rapidly and with remarkable success due to a variety of production processes, device engineering and material inventions. Nevertheless, the investigation and advancement of electrode materials and processes, the adjustment of rear electrode structures and the innovation of active layer materials to improve photon capture efficiency and transmittance in the visible range all hold great prospects.

Keywords: Transparent organic photovoltaic cell; power conversion efficiency; average visible transmittance.

1. Introduction

Solar energy, as a renewable energy can be used in a wide range of scenarios and domains. As the field of green energy is booming, how to better exploit solar energy has become a focus. The utilization of solar energy is in two ways: photothermal conversion and photovoltaic conversion. To convert solar energy to electricity more efficiently, solar cells have been developing rapidly. Organic photovoltaic cells (OPVs) are valued because of plenty advantages, especially the transparency. Transparent organic photovoltaic cells (TOPVs) can be applied to many scenarios of office windows, car windows, and electronic display screens and so on to provide electricity. OPV (containing TOPV) is composed of five layers: two electrodes on both sides, electron transport layer (ETL), hole transport layer (HTL), and the active layer which made by n-type semiconductor as the accepter and p-type semiconductor as the donor in the middle. The working mechanism of OPV is that the active layer absorbs photons to produce excitons. Excitons separate into electrons and holes at the interface of the donor and the accepter. Then it completes the charge transfer at ETL and HTL and the charge collection at the electrodes to conduct electricity. Due to the different active layers, the existing OPVs are divided into two types: bi-layer or multi-layer heterojunction organic photovoltaic cells and bulk heterojunction organic photovoltaic cells (BHJ-OPVs). Heeger et al. (1995) first mix donor MEH-PPV and accepter C60 and propose the structure of internal donor-accepter heterojunction which is the current BHJ [1]. BHJ-OPV has higher power conversion efficiency than the other one. Because it has a larger contact area of the donator and accepter, which can form countless tiny p-n nodes. It also reduces the exciton diffusion distance that more excitons can reach the interface for separation. TOPV is a special kind of OPV. Its devices need materials with high light transmission. However, this causes that TOPV absorbs less visible light and has a negative impact on the efficiency. Most TOPVs have the structure of BHJ to improve power conversion efficiency. Existing TOPV ordinarily use ITO as the anode, silver as the cathode, zinc oxide doped tellurium oxide as the ETL, PEDOT: PSS or molybdenum trioxide and other metal oxide as the HTL and PTB family polymers mixing with fullerene derivatives as the active layer. All the above materials have thin thickness and high visible transmittance. The preparation process of the active layer of TOPVs mainly includes: spin-coating, blade-coating and roll-to-roll coating (slot-die coating). Roll-to-roll coating can be used for large-scale preparation, which is most widely used method. At present, TOPVs are still in the research and development stage. Commercial promotion has not been achieved. This article will review the limitations and breakthroughs of TOPVs. It is supposed to point the direction for future research.

The two most significant characteristics about the development of TOPVs are efficiency and light transmittance. There are two appropriate and authoritative figures to measure these two properties: power conversion efficiency (PCE) and average visible transmittance (AVT). This review focuses on the optimization of PCE and AVT. It first reviews the idea of optimizing PCE and AVT separately. For the optimization of PCE, it proposes inserting photonic crystals and using fluorinated polymer materials. For the optimization of AVT, it proposes using carbon nanotubes. Because the electrodes in TOPVs is transparent, the photons cannot be reflected back to BHJ for reabsorption. This causes higher AVT but lower PCE. Therefore, for TOPVs, PCE and AVT are negatively correlated in most cases. The article then reviews the optimization of balancing PCE and AVT in using silver nanowire electrodes and antireflection coatings. The minimum ATV for TOPVs as a window is 25% [2]. It is significant to have a logical review of balancing PCE and AVT to create better OPVs. This review summarizes some outstanding achievements of previous research and the common reasons behind them. The review also indicates the gap and challenges in the research field of OPVs. It can put forward some ideas and directions for optimizing PCE and AVT of TOPVs for the research in the future.

2. The Optimization of Power Conversion Efficiency (PCE)

Efficiency is crucial to the TOPVs and determines the application value of them. PCE is an authoritative quality factor for evaluating efficiency of TOPVs. In this section, two main concepts of utilizing photonic crystals as the rear electrode device and fluorinated polymers as the active layer materials are proposed to optimize PCE of TOPVs.

2.1. Photonic Crystal

Photonic crystal (PC) is a kind of rear electrode technology. One-dimensional photonic crystals (1DPCs) mainly include distributed Bragg reflectors (DBRs) and dielectric mirrors (DMs). They are generally deposited onto the metal electrodes. 1DPCs are composed of multi-layer dielectric films with different refractive index. When the energy of the photons in the DBRs or DMs is smaller than the band gap of the PCs, they are reflected into the BHJ. The principle of their work is to use the interference of multi-layer dielectric films and reflect infrared and ultraviolet light selectively into the BHJ for reabsorption. PCs can improve PCE while keeping AVT basically unchanged or slightly improved.

Xia et al. [2] report a kind of PC which structure is MoO_3/LiF/MoO_3/LiF. After the calculation by transfer matrix method (TMM), it shows that compared with single-layer PC, double-layer PC can enable TOPVs to obtain higher PCE under the same visible light transmittance (VLT) conditions (Fig. 1). This implies that PC have considerable positive impacts on the improvement of TOPVs. When the TOPV is fabricated with PTB7-Th: IEICO-4F as the active layer, PCE can reach nearly 11% PCE and 23.7% AVT.

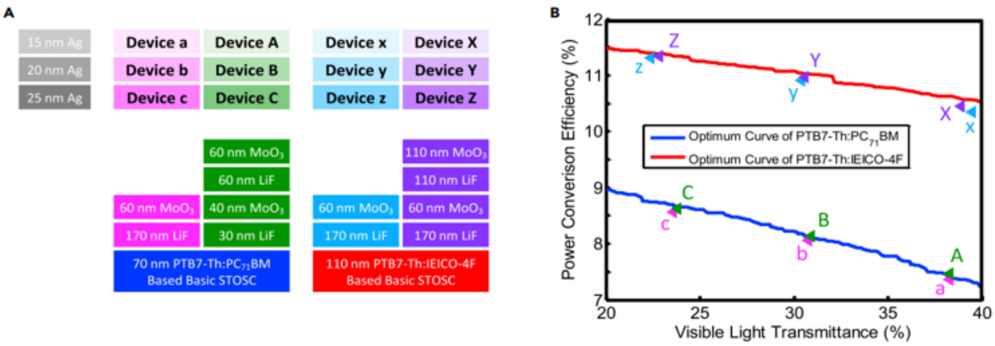

Fig 1. (A) TOPV devices selected after TMM.
(B) Simulated PCE-VLT coordinates of the selected devices [2].

Betancur et al. [3] reveal a kind of aperiodic PC that J_{sc} reaches the maximum value of 76.3% in the five-layer device structure as MoO_3 (102 nm)/LiF (136 nm)/MoO_3 (102 nm)/LiF (102 nm)/MoO_3 (102 nm). It can absorb infrared and ultraviolet light more efficiently. When PTB7:PC_{71}BM is used as the active layer, the maximum PCE is 5.6%, while the AVT is 30%.

2.2. Fluorinated Polymer Materials

Nowadays, PTB family polymers are widely used in the active layers as donors. Among them, PTB7 is the most popular one. Because of the impact of structure-property relationship, PTB7 (PTBF1) has the specific degree of fluoridation so that it can significantly improve PCE of TOPVs compared with other PTB family polymers. Research shows that introducing fluorine group on polymer backbone can make absorption blue shifted and reduce both HOMO and LUMO energy levels [4]. The introduction of fluorine influences the dipole moment. This is considered to be the vital reason of changing PCE. Lu and Yu [4] state that both ground state dipole moment and dipolar change between ground and excited state can affect PCE of TOPVs. PTB7 has the maximum dipolar change between ground and excited state and the highest PCE 7.40% compared with other polymers under the same conditions (Table. 1). At the same time, it explains the phenomenon that PTBF2 and PTBF3 introduce more fluorine while obtain lower PCE than PTB7 (PTBF1). The fluorine atom in thieno[3,4-b]thiophene (TT) in PTB7 (PTBF1) strengthens electron pulling ability of TT. However, the two fluorine atoms in benzodithiophene (BDT) in both PTBF2 and PTBF3 pull electron density in the opposite direction. This causes lower local charge transfer character. It also causes geminate recombination between holes and electrons in PTBF2 and PTBF3 and reduces the tendency to transfer to accepter. This indicates that it is decisive to increase the local dipole moment to improve PCE of TOPVs.

Table 1. Calculation of dipole moments and PCE values for several fluorinated polymer materials [4].

Polymer	μ_g(D)	μ_e(D)	$\Delta\mu_{ge}$(D)	PCE(%)
PTB2	3.60	6.37	2.96	5.10
PTB7	3.76	7.13	3.92	7.40
PTBF2	3.35	5.45	2.41	3.20
PTBB3	0.61	0.82	0.47	2.04
PBIT1	4.46	4.80	0.34	1.96
PBIT3	6.99	6.83	-0.16	0.47
PBTZ1	0.88	2.41	1.52	3.46
PBTZ2	1.92	1.48	-0.44	0.29

Carsten et al. [5] also report that due to the existence of two adjacent TT units in PTBF2 and PTBF3, it causes the internal polarization effect. This leads to the minimization of the internal dipole moment. It shows that PBB3 has the fastest recombination rate of the charge-separated (CS) species at the same time. Therefore, the degrees of charge separation in PBB3 and PTBF2 are very low. It proves the importance of local dipole moment and net dipole moment to the PCE of TOPVs.

3. The Optimization of Average Visible Transmittance (AVT)

The exceptional characteristic of TOPV is its high transmittance, which surpasses other photovoltaic cells. This section delves into the optimization of the AVT, which is an authoritative quality factor used to assess the transmittance of TOPV. It focuses on the remarkable role of carbon nanotubes in enhancing AVT.

Carbon nanotubes (CNTs) have a one-dimensional tubular structure which possess excellent conductivity, flexibility and transparency. CNTs are used for transparent conductive films (TCFs). CNTs are mainly composed of hexagonal carbon atoms to form several layers to dozens of coaxial round tubes. The carbon atoms in CNTs are mainly sp^2 hybridization. The hexagonal mesh structure has a certain degree of bending, in which a certain sp^3 hybrid bond can be formed. Therefore, the chemical bonds of CNTs have both sp^2 and sp^3 hybrid states. These p-orbitals overlap with each other to form a highly delocalized large π bond outside the carbon nanotube graphene sheet layer. The large π bonds on the outer surface of CNTs are the chemical basis for the non-covalent bonding of CNTs and some large molecules with conjugate properties. CNTs include single-walled carbon nanotubes (SWCNTs), double-walled nanotubes (DWCNTs) and multi-walled carbon nanotubes (MWCNTs). CNTs have three features: unideal carrier concentration and superior mobility, anisotropic electrical conductivity, and sensitivity to the environment [6]. Due to the high visible transmittance which is conductive to the AVT, CNTs, graphene and their related derivatives have gradually become ideal transparent electrodes of TOPVs.

Compared with MWCNTs, SWCNTs and DWCNTs are more widely used for TCFs od TOPVs due to they have less inner walls in the films so that have less non-current carrying voids which cause higher electrical conductivity. Jeon et al. [7] report the use of SWCNT as the transparent top electrode and p-doped in two methods, HNO_3 via sandwich transfer and MoO_3 via bridge transfer. Sandwich transfer has a higher PCE as 4.1% than 3.4% PCE of bridge transfer. When AVT reaches 90%, PCE drops to 3.7% under sandwich transfer with the structure ITO/ZnO/PTB7:$PC_{71}BM$/MoO_3/CNT. High AVT is indeed obtained at the expense of PCE. But to some extent, this also reflects the huge advantages and potential of CNTs in improving AVT.

Nair et al. [8] show that the opacity of graphene is defined by the fine structure constant and decreases linearly with the increase of the number of carbon atomic layers. Each layer absorbs 2.3% on the visible spectrum. SWCNTs have a porous network structure which can acquire superior AVT. To achieve the same light transmittance, the thickness of graphene needs to be much thinner than that of SWCNTs. In addition, experimental studies show that dry processing-based SWCNTs generally have higher conductivity than wet processing-based SWCNTs under the same AVT [6].

Conductive polymers doped in carbon nanostructures can also be used for transparent electrodes as new perspectives. Salvatierra et al. [9] state that polyaniline doped carbon nanotube (PANI: CNT) films can be used as organic photovoltaic device replacing ITO. PANI: CNT film has the best transmittance of 89% at 550 nm and high AVT with 2.27% PCE which is better than ITO, FTO and CNT. At the same time, PANI: CNT also has high flexibility and excellent mechanical stability.

4. The Balance between High PCE and AVT

PCE and AVT are negatively correlated in most conditions because high transparency results in absorbing less light and reducing efficiency. Therefore, it is indispensable to summarize the methods of balancing PCE and AVT. The following directions are offered to address the issue.

4.1. Silver nanowire electrode

Silver nanowires (Ag NWs), as exceeding conductive and flexible metal nanowires, are considered among the most promising materials for replacing traditional ITO transparent electrodes. The nanoscale dimensions of Ag NWs render them high transparent, concurrently ensuring remarkable efficiency. The forms of Ag NWs are various which can adapt to different conditions, such as very long silver nanowires (v-Ag NWs) and percolative network form Ag NWs. Ag NWs as the top electrodes, the main production methods are spray coating, spin coating, drop coating and lamination. In summary, Ag NW electrode is a device that enables TOPV to have both high PCE and AVT.

Ji et al. [10] research and show the highest 5.01% PCE and 50.3% AVT with the structure of ITO/ZnO/Polymer: PCBM/PMA: PEDOT: PSS/Ag NWs which have excellent overall performance. It is worth noting that the fully coated semitransparent devices with doctor-blade-coated functional layers and spray-coated Ag NW top electrodes of this TOPV performs much better especially in AVT than the TOPV based on thermally evaporated MoO_3/Ag/MoO_3 thin film electrodes (5.77% PCE and 19.5% AVT). This underscores the exceptional ability of Ag NW electrodes in reconciling PCE and AVT.

In addition to the independent utilization of Ag NWs as electrodes, the use of Ag NWs doped with organic and inorganic materials is also potential. Huang et al. [11] report the TOPV using a ferroconcrete-like AZO@silver nanowire (Ag NWs)@AZO (AAA) as the back stretchable transparent electrode (STE) have 12.83% PCE and 26.7% AVT. The full-solution-processed TOPV with thermoplastic polyurethane-embedded Ag NWs (TPU@AgNW) as the front STE sets a record as 10.9% PCE. After 500 cycles of 10% stretch-release, it can still maintain 76.5% of initial PCE which possesses outstanding tensile and mechanical stability.

4.2. Antireflection coating

Antireflection coatings (ARCs) are typically deposited on the surface of TOPVs which possess the ability to enhance transmission efficiency while capturing photons more effectively, thereby contributing to the advancement of both PCE and AVT. Organic ARCs reduce reflection by absorbing light and inorganic ARCs act by phase shift cancelling at specific wavelengths which are based on refractive index, film thickness and other parameters. Additionally, as a rear electrode device, ARC exhibit superior performance in improving AVT compared to PC.

Liu et al. [12] design an aperiodic bandpass filter (ABPF) as an ARC. With the structure of $(LiF/TeO2)^4$/glass/ITO/PEDOT: PSS/PM6: BTP-eC9:L8-BO/PDINN/Ag/ABPF, AVT can reach 78.69%. Compared with TOPV without ABPF, the TOPV with ABPF increases by 60% on AVT. When the composition ratio of PM6: BTP-eC9:L8-BO is 0.8:1:0.2 and the thickness of active layer is 75nm, the TOPV possesses 11.44% PCE and 46.79% AVT. In 140nm BHJ-based devices with ABPF, PCE achieves a record height of 15.10% with over 25% AVT which embodies the excellence of ARCs in optimizing PCE and AVT.

Lee at al. [13] have a remarkable ingenuity in using MoO_3 for both HTL and ARC. The TOPV has 5.74% PCE and 60% AVT with the device structure of ITO/ZnO/DPP2T: IEICO-4F/MoO_3/Ag/MoO_3. Due to the interference effect by alternating a low refractive index material and a high refractive index material, depositing a MoO_3 layer as the ARC onto MoO_3/Ag can improve AVT of the TOPV which is ingenious and convenient. In addition, sacrificing part of AVT can achieve PCE maximum as 9.13%.

5. Conclusion

The fabrication process of TOPVs is uncomplicated, cost-effective, and environmentally friendly. Their high transparency and lightweight properties render them suitable for a broad range of applications. TOPVs can replace large areas of glass in life scenes to provide electricity, and can be used in green energy vehicles, commercial large screens, and floor-to-ceiling Windows in high-rise buildings. In this review, it provides the optimization of PCE in utilizing the rear electrode of PC and the active layer of fluorinated polymer materials. Furthermore, the CNT electrode is suggested from the perspective of enhancing AVT. These two sections are not separate. On the contrary, they are interconnected and serve to enhance one another. Depending on the application scenarios, the requirements for the efficiency and transmittance (PCE and AVT) of TOPVs are different. It is approved to sacrifice part of PCE to meet the high AVT or lower the AVT to achieve a higher PCE, and then adopt different strategies accordingly. Moreover, the optimization ideas of PCE and AVT supplied in this review are aimed at different devices of TOPVs. Therefore, by combining PCE or AVT optimization schemes for different devices, TOPVs can achieve overall outstanding PCE and AVT. Apart from that, this review proposes the optimization of balancing PCE and AVT by using Ag NW electrode and ARC at the end, which also can guarantee the general performance of TOPVs.

In addition to the optimization strategies discussed in this review, a number of effective approaches can be employed to enhance the performance of TOPVs. These include but not limit to utilizing materials with weak visible light absorption and strong near-infrared absorption materials as the active layers, adjusting the ratio of donor and acceptor in the active layer, optimizing the electrode thickness, and implementing optical microcavity structure. TOPVs exhibit significant potential value in energy utilization and economic development. For electrode, rear electrode, ETL, HTL, and active layer, significant advancements can be made to optimize device structures, invent novel materials, and enhance production techniques, which would ultimately improve both PCE and AVT of TOPVs in the future.

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