

Enhancing Stability and Efficiency of Single-Material Organic Solar Cells with Block Copolymers

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Abstract: With the increasing demand for energy in production and the depletion of traditional energy sources, the urgency of exploiting clean energy is growing rapidly. Among them, building suitable organic solar cells (OSCs) by using block copolymers (BCPs) as materials of a photoactive layer is a promising method to utilize solar energy. Nevertheless, due to the complex structure and uncontrollability of BCPs, many efforts still need to be made to improve the power conversion efficiency (PCE) and thermal stability of the equipment. This paper reviews the development of BCP-based OSCs in recent years, as well as the many challenges they face and the corresponding solutions, including precise control of the block structure, the application of efficient charge transport principles, and optimization of active-layer film morphology. The paper takes the BCP-based single-material OSCs (BCP SMOSCs) as an entry point and provides a detailed analysis of the PCE and stability improvement schemes when the BCP is used as the light-active layer material, in order to provide a reference for the efficient utilization of photovoltaic products and the exploitation of renewable resources.

Keywords: single-material organic solar cells, block copolymers, power conversion efficiency, thermal stability, active layer

1. Introduction

Currently, considering the protection of the ecological environment, the demand for traditional energy sources such as natural gas is falling. In 2022, the world's natural gas consumption was 394 trillion cubic meters, with a year-on-year growth rate declining from 5.3% in the previous year to -3.1% [1]. It is of great significance for humans to rapidly develop a renewable energy conversion system with solar energy as the ultimate natural energy source [2]. The available solar energy is several thousand times greater than the world's energy consumption of about 9,000 Mtoe (million tons of oil equivalent) in 2000.

Due to the growing demand for solar energy, more attention has been paid to solar cells. Compared to the inorganic solar cells from c-Si, organic solar cells have better transmission properties and greater development potential. OSCs have seen progress through new organic π -conjugated materials. Donor-acceptor (D-A) polymers with fused rings in a D-A motif and controlled backbone orientation offer high charge carrier mobilities and wide absorption ranges for high-performance OSCs. Block copolymers, which are composed of two or more chemically distinct polymer chains linked together,

also have the excellent properties above [3]. Therefore, by conducting in-depth research on BCPs, suitable nanostructures can be more conveniently built and applied to the OSCs.

Although OSCs based on BCPs offer the potential for more cost-effective, tunable solar technologies, there are still some deficiencies in charge transport efficiency and in-use stability. In terms of charge transport efficiency, the key problem is how to improve the PCE of OSCs. In-use stability can be divided into two aspects: operation stability and thermal stability.

This paper, through a method of literature review, focuses on illustrating the mechanism of improving PCE and thermal stability by proposing some innovative polymer designs and treatment technology, and sequentially provides new insights for the preparation of high-performance OSCs in terms of structural design, especially in the short-range structure and condensed state structure of the macromolecular chain.

2. Basic Principles of organic solar cells

2.1. Photovoltaic properties of semiconductor materials

OSCs utilize the properties of semiconductor materials to convert solar energy. They operate on the basis of bandgap characteristics and the low-energy π - π^* transition for efficient light absorption [4]. In terms of photoelectricity conversion, the working principle of basic donor-acceptor (D-A) OSCs can be summarized into four steps, as shown in Figure 1. First, organic materials absorb sunlight to generate excitons. Second, excitons diffuse and dissociate into electrons and holes at the donor interface. Third, carriers migrate to the anode and cathode due to the electric field. Finally, a loop is formed for power output.

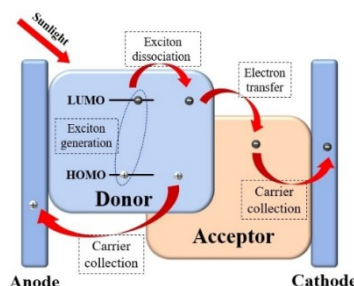


Figure 1: Working principle of basic donor-acceptor OSCs [4].

2.2. Different kinds of organic solar cells

According to the composition of the active layer, OSCs can be divided into several types. Single-material OSCs (SMOSCs) have electron donors and acceptors covalently bonded in the active layer. They have a metal electrode with limited conduction because its interior structure is mostly composed of polymer. Due to recombination in the same material layers, they have low PCE (about 4%) and aren't widely used in practical applications. When it comes to double-layer heterojunction OSCs, they use D/A sensitization at the heterojunction. However, due to the small exciton diffusion length, an active region is limited to about 10 nm. Thicker layers cause a masking effect, which also leads to a low PCE. To increase efficiency, the entire incident light must be absorbed in this thin active layer, which is difficult. In order to solve this issue, there come the bulk-heterojunction (BHJ) OSCs. BHJ OSCs have an active layer with a mixture of two materials forming a D-A system. Excitons are easily separated and diffused here to improve the PCE to 20%, but the challenge is the unstable morphology [5].

2.3. The polymer solar cells

In order to further improve the PCE and thermal stability of OSCs, it was discovered that using polymers as the internal structure of the cells, especially with π -conjugated materials, can be effective. In the early stages of research, the active layer was mainly made using organic pigments and dyes, resulting in dye-sensitized solar cells. As the research progressed, the active layer was made using a solution process of polymer π -conjugated materials and small molecule π -conjugated materials.

2.3.1. Dye-sensitized solar cells (DSSCs)

The dye-sensitized solar cell (DSSC), the third-generation solar cell introduced in 1991 by O'Regan and Gratzel, is promising in the OSCs field. It has advantages like rich raw materials, low cost, simple technology, non-toxic production, and a good trend of large-scale production. DSSCs are composed of organic (natural dyes) and inorganic (synthetic dyes) materials. In the aspect of organic materials, DSSCs are mainly synthesized from diverse parts of plants, such as leaves, fruits, petals, and bark. Among them, the most common organic dyes extracted are anthocyanin, chlorophyll, and flavonoids. For stability and efficiency, metal complex-based dyes are superior, but natural dyes are comparable in accessibility and environmental protection aspects [6].

The DSSC consists of five parts, as shown in Figure 2: the substrate or counter electrode (CE) allowing incident photons, the semiconductor acting as an electron-hole pair generator, the dye or sensitizer, a light-absorbing material being able to be extracted from plants or chemically synthesized, the electrolyte serving as a redox coupling agent for oxidation and reduction, and the counter electrode or back electrode supporting the front electrode (working electrode).

The dye-sensitized solar cell can achieve great PCE. Its operation involves several steps, as shown in Figure 2. (1) Sunlight excites electrons in dye molecules, transferring them from the ground state to the excited state. (2) These electrons are injected into the conduction band of the semiconductor. Meanwhile, the dye molecules are ionized and then accept electrons from the redox coupling agent, thus being reduced. (3) Electrons in the semiconductor conduction band move to the conductive glass interface and then to the external circuit. And (4) the oxidized state dye diffuses to the opposite electrode, gains an electron, and regenerates the reductant.

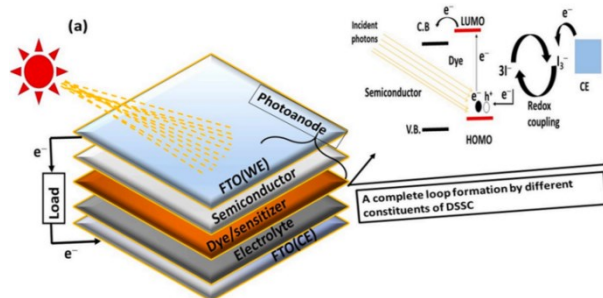


Figure 2: The working principle and components of DSSCs [6].

2.3.2. π -conjugated polymers for OSCs

π -conjugated polymers have the advantages of film-forming capacity and film stability over small molecules and have become a key material for organic solar cells. Using conjugated polymers as electron donor materials to bond with acceptors, the PCE is 8-10% [7].

In terms of device design, π -conjugated polymers are mainly used in the active layer of BHJ solar cells. The donor material in the active layer is made from π -conjugated polymers, while the acceptor material uses fullerene derivatives. The two are blended to form an active layer sandwiched between

the indium tin oxide (ITO) positive electrode and the low-workfunction metal negative electrode. On the ITO-positive electrode, a layer of poly (3,4-ethylenedioxythiophene) /polystyrene sulfonate (PEDOT/PSS) is also attached.

2.4. The BCP-based organic solar cells

BCP, due to its unique properties, such as self-assembly characteristics, can effectively connect various organic segments and is considered a promising material for preparing organic photovoltaic devices. For SMOSCs, their best PCE is far behind that of donor-acceptor BHJ OSCs (10% vs. 19%). However, recently SMOSCs based on BCP have broken through this efficiency bottleneck - the best PCE of SMOSCs using PM6-b-PY-IT has reached 18%, exceeding the 16.22% of the corresponding binary all-polymer counterparts [8].

2.4.1. The structure and principle of BCP SMOSCs

In traditional BHJ active layers, the electron donor is usually a conjugated molecule or polymer, while the electron acceptor is a fullerene derivative. As the BHJ concept continues to evolve, materials with covalently bonded electron-giving and electron-withdrawing groups have begun to attract increasing attention. Functional materials such as block copolymers, bicomponent copolymers, and small-molecule dyads are designed and successfully prepared for SMOSCs. The working mechanism of BCP SMOSCs is different from that of typical BHJ OSCs. In BCP, functionalized donor and acceptor segments chemically bond to each other, so there may be intra-chain charge separation and transport [9]. In this case, BCP can self-assemble into ordered microstructures and remain relatively stable while promoting charge carrier transport to improve PCE.

Based on their chemical structure and development sequence, BCP SMOSCs can be mainly divided into three generations, as shown in Figure 3. The first generation of BCP has electron donor and acceptor segments as substituents attached to a flexible main chain, but the PCE is poor, only 0.5%; The second generation of BCP has the donor segment as a part of the conjugated main chain while the acceptor segment remains as a substituent, which enhances the charge transport and raises the PCE to about 2%; the third generation of BCP uses a fully conjugated main chain - the acceptor and donor segments are both on the main chain, which raises PCE from 3% to 15%.

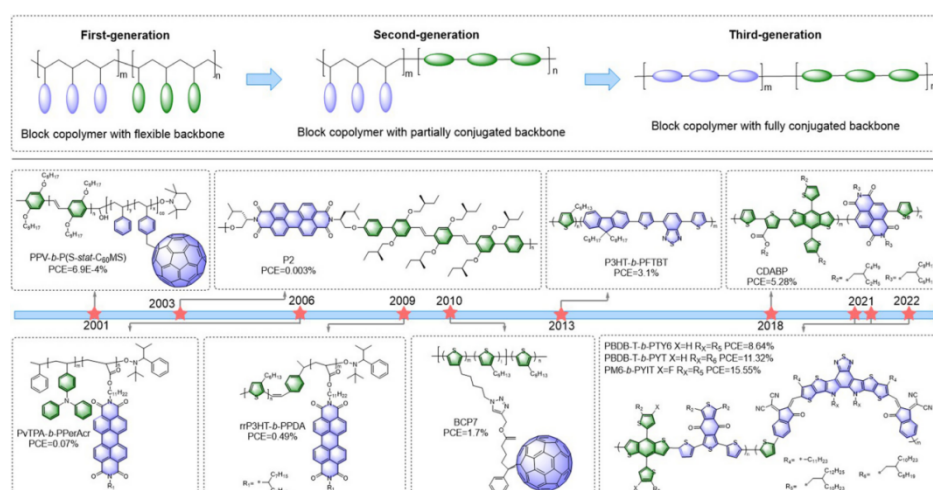


Figure 3: The short-range structure of three generations of BCP molecular chains and the major progress of three generations of BCP SMOSCs in recent years [9].

It is obvious that significantly improving the PCE of the third-generation BCP SMOSCs often requires the combination of relevant post-treatment methods. For example, for PM6-b-PY-IT, a two-step annealing method combining solvent vapor annealing (SVA) and thermal annealing (TA) can increase the photon utilization rate and charge transport efficiency, thereby improving device efficiency, optimizing crystallinity and phase distribution [10].

2.4.2. The challenges that BCP SMOSCs needs to address

Despite the fact that designing and synthesizing BCPs with suitable architectures as the active layer material for SMOSCs has been proven to significantly enhance their PCE and even make them superior to the corresponding properties of the binary full-polymer counterparts, there are still some challenges in improving the PCE and thermal stability of BCP SMOSCs.

First of all, the molecular weight of the polymer has an important influence on its photovoltaic and mechanical properties, such as the molecular interfacial interaction between polymer chains increasing with the molecular weight. However, the solubility of the polymer decreases with excessive molecular weight, which affects its mechanical properties.

For the SMOSCs film, there are not only contact-type D/A mixed interfaces, but also chemically bonded D/A interfaces - the chain interconnections between the electron donor and acceptor provide an additional path for charge transfer. However, the reaction speed at the interface is fast, and the contraction strength is also large, which is not conducive to the dissociation of excitons and the transport of charges.

Moreover, molecular stacking and active-layer film morphology are important factors affecting OSC performance, and the photovoltaic performance is greatly affected by the stacking pattern of the groups. Some post-treatment methods need to be used to improve the morphology.

In addition, in order to ensure the sufficient practicability of BCP-based OSCs, the efficiency and convenience of their large-scale production and preparation should also be considered. Most small-area OSCs are fabricated by spin coating, but the shape optimization methods developed from this, such as TA and SVA, are not suitable for large-area OSC manufacturing. In order to achieve large-scale industrial production, exploration of methods such as blade coating is needed, though the large-scale manufacturing process may cause PCE loss.

3. Methods for improving PCE and thermal stability of BCP SMOSCs

3.1. Precise control of the block structure

3.1.1. Ways to enhance PCE

SMOSCs based on conjugated block copolymers (CBCs) are increasingly popular due to their good stability and morphology. Nevertheless, the effect of assembly behavior induced by CBC block structure on device performance is still in the exploration stage. Heung-Goo Lee et al. synthesized a series of D18-b-PDMS with different D18 block lengths by sequential polymerization (Figure 4) [11]. They enhanced the intermolecular interactions by increasing the number-average molecular weight (M_n) of the D18 block, which improved the crystallinity and electrical properties. As a result, the PCE of OSCs was increased from 15.1% for D18L-b-PDMS to 17.3% for D18H-b-PDMS.

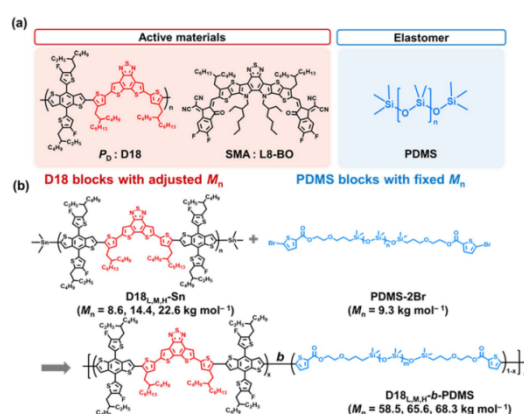


Figure 4: (a) Molecular structures of the active materials and the elastomer. (b) Synthetic route of $D18_{L,M,H}$ -b-PDMS [11].

However, this does not mean that the length of the D18 block can be increased indefinitely in order to raise the PCE. Yujun Cheng et al. synthesized a series of D18-b-PYIT conjugated BCPs with different block structures by two-pot polymerization (Figure 5) [12]. The main difference is the length of the D18 block. Due to the outstanding capacity of adjusting the self-assembly of CBCs and improving optoelectronic properties, the D18(40)-b-PYIT-based device with a moderate block length achieved a high PCE of 13.4%.

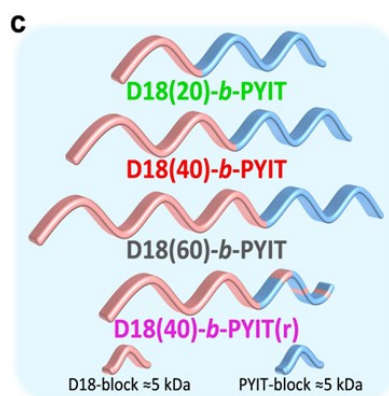


Figure 5: Short-range structure of $D18(20,40,60)$ -b-PYIT [12].

3.1.2. Ways to improve thermal stability

In addition to improving the PCE of solar cells, the main principle of using block control to improve its thermal stability is providing a thermodynamic driving force for mesoscale structure formation. Zixuan Guo et al. synthesized the PTB7Th-b-PNDI donor-acceptor BCP via sequential Stille reactions. Based on the principle of promoting favorable phase separation and molecular packing in the active layer, as well as maintaining the crystallinity, they added 5% by weight of this BCP into the active layer. It effectively reduced 30% of the thermal degradation of photovoltaic devices under thermal stress at 110°C for 168 h, enhancing the thermal stability [13]. The main reason for this phenomenon is that the added PTB7Th-b-PNDI effectively suppresses the interaction between the aluminum electrode and the photoactive layer, reducing the chemical degradation of the active layer surface.

Another feasible idea to increase the thermal stability of equipment is to prepare and apply high-performance interface compatibilizers. It can improve the interface compatibility between the photoactive components, thus maintaining the stability of the device structure under thermal stress.

With further research on the efficiency of OSCs, unfavorable molecular interactions at the interface between donor and acceptor blocks have also been found to be an important factor affecting PCE. In this direction, Tan Ngoc-Lan Phan et al. have developed a new conjugated block copolymer PBDB-T-b-PY5BDT [17]. By introducing the same benzodithiophene (BDT) unit into the donor and acceptor blocks, the polymer enhances the compatibility between blocks and forms a good intermixing domain, which is conducive to charge generation. At the same time, adequate percolation and high crystallinity ensure efficient charge transfer, enabling organic solar cells based on the polymer to achieve a high PCE of 10.55%.

In addition, the different synthesis methods of BCPs are also one of the potential influencing factors of PCE. There are two typical synthesis methods currently, one is direct synthesis, in which the monomers of polymer A are first copolymerized to form polymer A, and then the monomers of polymer B are copolymerized with polymer A to form a BCP composed of A and B. The other method is sequential synthesis, in which the monomers of A and B are copolymerized separately to form two polymers, and then the two polymers are copolymerized to form a BCP. Bin Li et al. comparatively studied the PBDB-T-b-PYIT synthesized by direct synthesis (d-BCP) and sequential synthesis (s-BCP) (Figure 8) [18]. The d-BCP has a purer phase composition, enhanced molecular ordering, and higher intramolecular charge transfer efficiency. The intramolecular hole transfer rate of d-BCP is about an order of magnitude higher than that of s-BCP, and the charge transfer and the exciton dissociation are faster, thus achieving a high PCE of 15.02%.

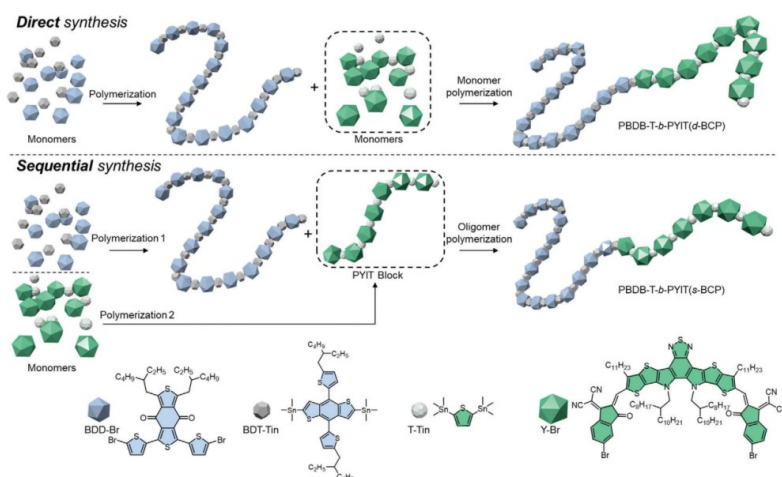


Figure 8: Schematic diagram of direct and sequential synthesis of PBDB-T-b-PYIT [18]

3.2.2. Ways to improve thermal stability

The biggest difference between BCP SMOSCs and BHJ OSCs is that the light-active layer in the former is formed by bonding the acceptor and donor in a molecular form, while the latter uses a blending method to transfer charges between the acceptor and donor. Since some small molecule acceptor materials used in BHJ OSCs may affect thermal stability due to their tendency to diffuse easily [19], using the form of BCP to fix these small molecules on the BCP and form a macromolecular material is a feasible solution to improve thermal stability of the OSCs. In this field, Bin Li et al. designed the PM6-b-PTY6 as a compatibilizer for non-fullerene OSCs [20]. Regarding thermal stability, adding 5 wt% of PM6-b-PTY6 significantly improves the morphological thermal stability of the PM6:Y6 system. After thermal aging at 80°C for 550 hours in a nitrogen atmosphere, the device with this compatibilizer can maintain a PCE of 0.86 of the initial value, while the PCE of the control device without the compatibilizer drops to 0.71 of the initial value. This is because the

compatibilizer suppresses the diffusion of Y6 molecules and reduces the impact of environmental factors on the blend morphology, thereby enhancing the long-term thermal stability of the battery.

3.3. Optimization of active-layer film morphology

In terms of enhancing the performance of BCP SMOSCs, improving PCE is the central goal of active-layer film morphology optimization. This can be achieved in the two following ways.

3.3.1. Adjustment of molecular stacking pattern

The different film morphologies can reflect the differences in molecular stacking patterns in the photoactive layer of BCP SMOSCs, thereby affecting the optical properties of the active layer. Two typical stacking modes are A-to-A type J-aggregation and A-to-D type J-aggregation. An ideal stacking pattern can simplify the diffusion and dissociation pathways of excitons, thereby improving PCE (Figure 9).

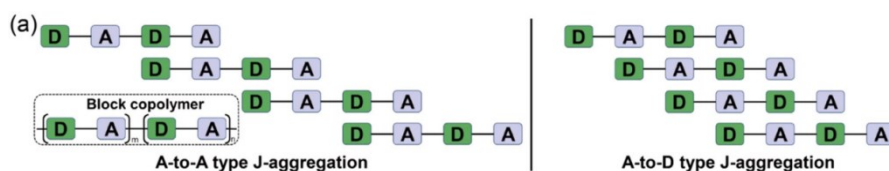


Figure 9: Schematic diagram of two typical stacking patterns (A-to-A type J-aggregation and A-to-D type J-aggregation) [9]

In this context, Xinrong Yang et al. synthesized three chlorine-substituted position isomers and introduced CBCs [21]. The change in chlorine position affects the molecular stacking and charge transport. In the three isomers, PB-b-PY-2 has the most ordered molecular stacking, with a π - π stacking distance of 3.81 Å and the highest crystal coherence length (CCL) of 15.408 Å, resulting in higher charge mobility and better balance ($\mu_h/\mu_e = 1.01$), thereby enabling the highest PCE (13.42%). Na Yeon Kwon et al. used TEM images to observe the internal morphology of the light-active layer directly [22]. They prepared polymer solar cells based on cross-linked P (OXBTT-co-NDI2T) films, which showed higher PCE than those based on cross-linked blend films. From the TEM image analysis, it was found that the photo-crosslinking reaction effectively reduced the phase separation between hydrophilic P (OXBTT) and hydrophobic P (NDI2T), forming a more uniform film morphology and a complete network structure.

3.3.2. Reasonable use of post-treatment methods

At present, the traditional film post-treatment method is a two-step annealing method (first SVA, then TA), which is an effective method to anneal BCP films and tune the self-assembly of BCPs at low temperatures. Linji Yang et al. adopted this two-step annealing method to treat the films of PM6-b-PY-IT-based OSCs [10]. SVA increased the crystallinity of the film, reduced the degree of phase separation, formed a more ordered molecular stacking, and promoted charge transport, thereby increasing the J_{sc} and fill factor (FF) and achieving an efficiency of 15%. Based on this, Shangfei Yao et al. made technological improvements. They changed the previous method to a two-in-one annealing strategy (solvent vapor thermal annealing, SVTA) to regulate the active-layer film morphology for OSCs based on PM6-b-PY-IT [8]. Compared with SVA + TA, it can form a more favorable interpenetrating network morphology with a smaller domain size, which is beneficial for improving both the charge transport efficiency and the FF, thus achieving a higher PCE of 16.08%.

4. Conclusion

In summary, in order to promote the wider application of OSCs, enhancing their PCE and thermal stability is a complex and ongoing research topic. This paper summarizes some advanced research results in recent years in terms of performance enhancement through comparative and analytical synthesis. For example, in the part explaining the efficient charge transport principle, the three-step research on the study of whether the active layer components are covalently bonded, the bonding orientation and form, and whether a compatibilizer is designed in the segment was conducted in order to highlight the continuous refinement and deepening of research in charge transport, thus better showcasing the iterative optimization of the idea. In addition, in analyzing the precise control of block structure and optimization of active-layer film morphology, the paper respectively compares two studies on the length of the D18 segment and two different annealing methods. However, there are still some unsolved or incomplete problems in this research topic, which need further research and exploration. In terms of OSC types, this paper only focuses on the performance enhancement analysis of BCP SMOSCs, but the BHJ OSCs are also one of the objects of extensive application and research, and their PCE enhancement has good exploration prospects. For the challenges of BCP SMOSCs application, the paper also mentioned that large-scale production requires preparation methods other than spin-coating, but since the theme of this paper is to enhance the internal performance of the device - PCE and thermal stability - it has not been analyzed in this aspect. In fact, the optimization of the preparation process should be the research focus after the performance optimization.

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