Emerging Approaches in Enhancing the Efficiency and Stability in Non-Fullerene Organic Solar Cells

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The past three years have witnessed rapid growth in the field of organic solar cells (OSCs) based on non-fullerene acceptors (NFAs), with intensive efforts being devoted to material development, device engineering, and understanding of device physics. The power conversion efficiency of single-junction OSCs has now reached high values of over 18%. The boost in efficiency results from a combination of promising features in NFA OSCs, including efficient charge generation, good charge transport, and small voltage losses. In addition to efficiency, stability, which is another critical parameter for the commercialization of NFA OSCs, has also been investigated. This review summarizes recent advances in the field, highlights approaches for enhancing the efficiency and stability of NFA OSCs, and discusses possible strategies for further advances of NFA OSCs.

1. Introduction

It has been more than 30 years since the first solar cell based on organic heterojunctions was reported by C. W. Tang. Different acceptors, including fullerene derivatives, inorganic nanocrystals, and more recently, small molecules, have been investigated as acceptor materials in OSCs. Although fullerene had dominated the field for over two decades, this situation has changed recently. Benefiting from the structural features, distinctive advantages, and great synthetic flexibility, fused-ring non-fullerene acceptors (NFAs) have been developing rapidly since 2015, enabling high power conversion efficiencies (PCE) approaching 18% (Figure 1a).

The fused-ring NFAs consist of three structural features, as shown in Figure 1a, including a ladder-type fused donor unit at the center, side-chains hanging out from the molecular plane, and two strong compact electron-withdrawing end groups. Independent modifications of these three parts provide diverse NFA molecular structures, thus enabling strong absorption in the visible and/or near infrared (NIR) regions, easily tunable energy levels, and finely tunable crystallinity. In addition, low voltage losses are a significant feature of NFA OSCs compared to their fullerene counterparts, contributing to the rapidly increasing PCE. At the same time, the operational stability of NFA OSCs has also been demonstrated to be promising, although further improvement is required to meet the standard for practical applications.

Along with the rapid development of materials and devices, the fundamental understanding of OSCs is also moving forward, though at a slower pace compared with that of material development and device engineering. One of the greatest impediments to developing a fundamental understanding of OSCs lies in the bulk heterojunction (BHJ) structure. The invention of BHJ, which is a milestone in OSC research, increases the interfacial area between the electron donor and electron acceptor materials, overcoming the issues of short exciton diffusion length, limited exciton lifetime, and charge separation that limits bilayer junctions. Together with these advantages, the BHJ structure also presents challenges. The multiple phases and complex interfaces bring about sophisticated hierarchical morphologies and complicated charge dynamics, which are difficult for experimental observation and control. In NFA-based systems, some of these challenges have even been manifested. The similarity in the element constitution makes it difficult to spatially distinguish the electron donor and electron acceptor materials. Additionally, the resemblance of energy makes the spectra indistinguishable for charge-transfer (CT) states and singlet excitons. Furthermore, OSC materials are updated so fast that the new materials can be distinct from previous ones, and the characteristics may be entirely different.

This review covers emerging approaches for enhancing the efficiency and stability of non-fullerene OSCs. We highlight that recent advances on non-fullerene OSCs result from the coordinated development of donors and acceptors; those who are interested in a comprehensive understanding of donor materials are referred to recent review articles on this topic. In this review, we mainly focus on the development of acceptors. We summarize new strategies for high short-circuit current ($J_{SC}$) and fill factor (FF). A variety of methods, for example, extended absorption and effective morphology control, are discussed. We also introduce design rules for low energy losses, especially by...
suppressing non-radiative losses. Moreover, we present the investigations on the degradation mechanism of non-fullerene OSCs, as well as the methods to further improve the stability. At the end of this review, we provide an outlook on the advantages of non-fullerene OSCs, along with opportunities for improvements, and suggest design strategies for highly efficient and stable OSCs for commercially viable photovoltaics. Previous estimations indicate that a high efficiency of 20% is possible for OSCs (Figure 1b). We hope that this review provides useful insights to help reach this target and improve the stability of OSCs.

2. Emerging Approaches in Enhancing Device Efficiency in NFA OSCs

2.1. Emerging Strategies for High \( J_{\text{SC}} \) and FF

In this section, we discuss the optimization strategies for \( J_{\text{SC}} \) and FF based on the structure-property relationship of NFAs and recently reported device engineering. In order to achieve an optimum \( J_{\text{SC}} \), it is important to extend the absorption to the NIR region, through either material design (narrow-gap materials) or device development (ternary or tandem devices). In addition, it is important to control the morphology of the active layer and understand the charge transport and charge recombination, which are also key factors affecting \( J_{\text{SC}} \) and FF.

2.1.1. Absorption Expansion for Enhanced Photon Utilization

An intuitive method to enhance the light absorption of the active layer is to increase the film thickness. However, due to the limited charge mobility of organic semiconductors, the charge recombination increases dramatically in thick films, leading to a decreased FF and even a reduced \( J_{\text{SC}} \) at the end. Here, we will discuss the narrow-gap molecule design and device engineering, including tandem and ternary strategies, to broaden the absorption of the active layer to the NIR region, where the density of photon flux is high in the solar spectrum.

Narrow-Gap Molecule Design: The energy levels and optical bandgaps of NFAs can be tuned by controlling the intramolecular charge transfer interaction between the donor moiety and electron-withdrawing end groups, according to the molecular orbital theory. To narrow the bandgaps of NFAs, one can enhance the electron-donating ability of the donor core using the following approaches (shown in Figure 2): 1) Extending the effective \( \pi \)-conjugation length. By inserting electron-rich oxygen atoms into the donor core, another strategy to extend the absorption is to introduce fluorine atoms onto the end group. For example, by extending the donor core from a five-fused-ring (e.g., ITIC) to a seven-fused-ring (e.g., IDCIC), the bandgap can be reduced from 1.70 to 1.59 eV by extending the donor core from naphthalene-based IHIC2 to a naphthodithiophene-based IOIC2 and then to a fused decacylic donor core based IDCIC, the absorption onset shows a redshift from 745 to 801 nm and to 853 nm, and the maximum extinction coefficient increases from 1.6 \( \times \) 10^3 to 1.8 \( \times \) 10^3 M^-1 cm^-1 and to 3.3 \( \times \) 10^5 M^-1 cm^-1. As a result, the \( J_{\text{SC}} \) of the corresponding devices increases from 16.1 to 19.7 mA cm^-2 and to 21.98 mA cm^-2.

2) Introducing electron-donating groups. For example, by introducing alkoxothiophene to ITDIC, the bandgap of IEICO is reduced to 1.34 eV by inserting electron-rich oxygen atoms into the donor core, a CO-bridged ladder-type NFA (CO-8DFIC) was reported, with a small bandgap of 1.26 eV and strong absorbance in the range of 600–1000 nm. Thus, OSCs based on CO-8DFIC achieved an impressive \( J_{\text{SC}} \) (28.20 mA cm^-2), which is the highest reported \( J_{\text{SC}} \) among OSCs. 3) Introducing quinoidal resonance structures. For example, by inserting thieno[3,4-b]thiophene into ITDIC, we obtain ATT-2, which possesses a narrow bandgap of 1.32 eV with the absorption onset redshifted to 940 nm and the maximum extinction coefficient improved to 2.0 \( \times \) 10^5 M^-1 cm^-1. As a result, the ATT-2 based OSCs showed improved \( J_{\text{SC}} \) for higher PCE.

In addition to enhancing the electron-donating ability of the donor core, another strategy to extend the absorption is to strengthen the electron-withdrawing capacity of the end group, resulting in a downshift of the lowest unoccupied molecular orbital (LUMO) energy level for redshift absorption and high \( J_{\text{SC}} \). Typically, halogen atoms (fluorine and chlorine) are introduced to the end group. For example, Hou and coworkers attached fluorine atoms onto the end group of IEICO and developed IEICO-4F with a narrow bandgap of 1.24 eV. Its absorption spectrum shows a redshift of approximately 75 nm compared with the original compound, IEICO. Although
the downshift of the NFA LUMO energy levels can reduce the bandgap for high $J_{SC}$. the open-circuit voltage $V_{OC}$ of the devices will also decrease concomitantly. Hence, balancing this tradeoff between $J_{SC}$ and $V_{OC}$ is a challenge when using this strategy. To compensate for the tradeoff between $J_{SC}$ and $V_{OC}$, it is essential to manipulate the energy levels of the donors so that they match well with those of acceptors for low energy losses. For example, IT-4F was developed by the fluorination of ITIC for extended absorption; in this case, the polymer donor (PBDB-T) also needs to be fluorinated (PBDB-T-SF) to match the energy levels of IT-4F. As a result, the PBDB-T-SF:IT-4F blend shows improved photocurrent (due to extended absorption) and maintains the photovoltage, yielding an improved PCE of 13.1%. In addition, the donor-acceptor1-donor-acceptor2 (D-A1-D-A2) copolymerization strategy was also adopted to achieve the low highest occupied molecular orbital (HOMO) energy level of the polymer donors. For example, the polymer PhI-fBBT obtains a deep HOMO of $-5.55$ eV, high hole mobility, and complementary absorption with IT-4F. As a result, the PhI-fBBT:IT-4F solar cell affords a high PCE of 13.31% with an impressive $V_{OC}$ of 0.94 V, a $J_{SC}$ of 19.41 mA cm$^{-2}$, and an FF of 0.76.

**Tandem and Ternary Strategy:** Benefiting from the great diversity of organic materials, tandem and ternary strategies can effectively broaden the absorption region of OSCs and enhance the external quantum efficiency (EQE). Yang and coworkers introduced a low bandgap polymer PDTP-DFBT in the back cell, broadening the absorption to 900 nm. This tandem device based on P3HT:ICBA and PDTP-DFBT:PCBM (the chemical structure is shown in Figure S3, Supporting Information) affords the first certified polymer solar cell efficiency of over 10%. With the development of NFAs, the absorption region of tandem solar cells can easily achieve a narrow-gap (>900 nm). In addition, the energy loss of each sub-cell is also reduced for enhanced $V_{OC}$. Thus, the efficiency of tandem solar cells has increased significantly in recent years. For instance, Forrest and coworkers used DTDCPB:C$_{70}$ as the front cell via vacuum thermal evaporation and PTB7-Th:BT-CIC as the back cell by spin-coating to prepare tandem solar cells. It has strong absorption in the range of 350–950 nm. The tandem solar cell afforded a PCE as high as 15%. Recently, a certified record PCE of 17.29% (Figure 3a) for a two-terminal monolithic solution-processed...
tandem OSC was achieved based on the front cell PBDB-T:F-M and the rear cell PTB7-Th:CO,8DFIC:PC71BM after careful calculation and model analysis. As shown in Figure 3b, the EQE of the tandem cell exhibits a high overall value of 72% from 300 to 1000 nm. The front cell mainly absorbs photons in the range of 300 to 720 nm with a maximum EQE of 76% at 560 nm, while the rear sub-cell shows a high EQE response in the range of 720–1050 nm. The high and balanced JSC of the two sub-cells originates from their complementary absorption and strong photon response.

Compared with tandem solar cells, ternary solar cells have the potential to realize the complementary properties of different materials within a single junction, which avoids the complex multi-junction stack structure and reduces the processing cost. In the J52:IT-M:IEICO ternary OSCs, IT-M has strong absorption in the range of 600–750 nm and it efficiently makes up the gap of J52:IEICO binary system in absorption. Thus, the ternary OSC obtained strong absorption in the range of 350 to 900 nm. In addition, efficient excitation energy transfer from IT-M to IEICO was also demonstrated. As a result, the ternary OSC achieved an enhanced JSC of 19.7 mA cm−2 after optimizing the component ratio, as shown in Figure 3c,d. Similarly, Zhang and coworkers applied the NFA, MeIC1, as the third compound to the PBDB-T:Y16 binary system and filled the dip at 670–730 nm in the absorption spectrum. Thus the PBDB-T:MeIC1:Y16 ternary solar cell affords a PCE of 14.11% with an enhanced JSC of 22.76 mA cm−2. Besides the charge transfer and energy transfer mechanism, the third component can also elevate the performance via morphology control which will be discussed in the next part of morphology control.

2.1.2. Morphology Control

A critical factor that affects the performance of a given donor/acceptor (D/A) system is the morphology of the active layer, which is strongly dependent on the molecular properties (e.g., solubility, crystallinity, miscibility, etc.), film processing, device configuration, etc. There are two phase-separation mechanisms for polymer blends; one is nucleation and growth phase separation, and the other is spinodal decomposition. When two components are not well mixed on the molecular scale, additional nanoscale phase separation forms. If the polymer blend phase separation can be effectively controlled, it will be an efficient way to assemble structures on the nanometer scale. However, the phase separation mechanisms for polymer blends may not be suitable for all small-molecule OSCs, due to the different phenomena observed in all small-molecule blends. For instance, the ZR1:Y6 blend affords a hierarchical morphology with a large domain size of approximately 70 nm. The phase-separation mechanism for all small-molecule OSCs remains unclear and hence requires further investigations. Since there have been previous reviews covering general methods for morphology control, we will focus on the phase separation and molecular stacking from the D/A structural point of view, as well as the latest strategy of morphology control.

Phase Separation: The phase separation in OSCs usually denotes the segmentation of the donor phase, acceptor phase, and donor/acceptor mixed phase in the formation of BHJ films. The degree of phase separation can be quantitatively analyzed by characterizing the domain size and domain purity. It is of
critical importance to control the degree of phase separation in OSCs. On one hand, the efficient generation of CT states requires reasonably small domain sizes (∼20 nm) due to the limited exciton diffusion length in organic semiconductors. On the other hand, high domain purity is necessary to enable efficient charge extraction and inhibit charge recombination. From the point of view of polymer-donor materials,[44,45] a typical approach for phase-separation control is based on a family of polymers that exhibit strong temperature-dependent aggregation (TDA) properties in solution. These polymers with strong TDA properties, for example, PffBT4T-2OD, have worked well in fullerene-based OSCs.[30,88] For NFAs, the polymers with slightly reduced TDA effects tend to work better. Among them, the most successful ones are PBDB-T and its derivatives (such as PM6).[22,35,43,89–92] It has been found that the π-π aggregation shoulder peak in the absorption spectra of PBDB-T gradually disappears and its absorption shows an obvious blueshift as the solution temperature increases. This distinct TDA property benefits the formation of nanofiber structures in the active layer and probably confines the nucleation and growth of the acceptors, thus achieving well-segregated donor/acceptor domains.[50] For example, in the PM6:Y6 system, obvious nanofiber structures with optimal phase separation can be found from atomic force microscopy (AFM) and transmission electron microscopy (TEM) images, contributing to efficient exciton dissociation and charge transport.[22] As a result, PM6:Y6 based OSCs gave a remarkable PCE of 15.7% with a high $J_{SC}$ of 25.2 mA cm$^{-2}$ and a high FF of 76.1%. It has been well accepted that the TDA properties of polymers can help the formation of nanoscale phase separation in the BHJ active layer, but the detailed underlying mechanisms require further systematic investigations.

In addition, the aggregation behavior of the polymer donor can also be tuned by side-chain engineering. As shown in Figure 4, the fibrillar structure of the neat polymer film can be effectively tuned by changing the atom that links the conjugated backbone and side chains.[93] PBT1-C forms narrower fibril widths (∼8.5 nm) than PBT1-O and PBT1-S, facilitating charge carrier generation and transport. As a result, PBT1-C based fullerene OSCs and NFA OSCs deliver high FF of 80.5% and 78.5%, respectively, due to the formation of an optimal interpenetrating network morphology.

As for all small-molecule OSCs, small-molecule donors with strong crystalline properties have also worked well with NFAs, providing another way to manipulate the morphology and induce well-segregated phases.[85,94,95] The small molecule donor, ZR1, possesses high crystallinity and thus enables the formation of the hierarchical morphology in the ZR1:Y6 blend.[85] On one hand, a certain number of ZR1 crystals and amorphous ZR1:Y6 intermixed regions within large ZR1-rich domains contributed to exciton dissociation in the BHJ. On
the other hand, the high crystallinity of ZRI facilitates excellent charge transport. Therefore, ZRI:Y6 based solar cells achieve an impressive PCE of 14.34%.

For NFA materials, the capacity of the formation of a favorable domain size is also important. Typically, highly crystalline organic semiconductors, widely used in transistors for high mobility, tend to form excessively large domains in the blend; whereas amorphous materials can generally gain small domains but suffer from low domain purity and hence poor charge transport. This dilemma has been efficiently solved by developing perylene diimide (PDI) oligomers with twisted structures or linearly fused-ring electron acceptors (FREAs) with aromatic backbone having sp3 carbon bridge atoms, such as the landmark material, ITIC.[31,67,89–98] In particular, the latter has shown excellent tolerance towards chemical modifications, enabling ITIC and its derivatives to be a series of high-performing materials after matching well with the crystallinity and miscibility of the donors.

We notice that, even among FREAs, the molecular structure also significantly impacts the domain size in the blend film. Zhan and coworkers reported a series of small bandgap FREAs (F6IC, F8IC, and F10IC) by gradually extending the donor core in the center.[99] Grazing-incidence small-angle X-ray scattering results show that the acceptor domain sizes in PTB7-Th/F6IC, PTB7-Th/F8IC, and PTB7-Th/F10IC blend films are 54.2, 18.5, and 14.8 nm, respectively. Therefore, PTB7-Th/F8IC and PTB7-Th/F10IC based solar cells achieved impressive J and 14.8 nm, respectively. Therefore, PTB7-Th/F6IC, PTB7-Th/F8IC, and PTB7-Th/F10IC blend films are 54.2, 18.5, and 14.8 nm, respectively. Therefore, PTB7-Th/F6IC, PTB7-Th/F8IC, and PTB7-Th/F10IC based solar cells achieved impressive JSC of 25.12 and 20.83 mA cm⁻², respectively, and are higher than PTB7-Th/F6IC based solar cells (18.07 mA cm⁻²), owing to their smaller domain sizes in favor of exciton dissociation and charge transport. Recently, Yang and coworkers found that the PBDT-TIDICT blend film is amorphous and does not exhibit obvious phase separation, because of the serious steric hindrance of the bulky phenyl attached to the main backbone of IDTIC for its poor crystallinity.[100] However, the PM6:IDIC blend film shows a large aggregation of IDIC and serious phase separation, due to the high crystallization of IDIC with alkyl chains. These two cases both show inferior PCE (6.41% for IDTIC and 12.02% for IDIC) due to poor charge carrier dissociation or transport. Then they attached the phenyl butyl to the molecular backbone and developed IDIC-C4Ph. It has moderate crystallization and achieves optimized morphology with PM6. PM6:IDIC-C4Ph solar cell achieved the best PCE of 14.04%. Hence, it is vital to carefully manipulate the molecular structure of NFA to optimize the phase separation in the active layer.

As discussed above, both donor and acceptor structures affect phase separation, since their intrinsic molecular structures determine their compatibility, which governs the phase separation of D/A blends.[101] From the perspective of quantitative analysis, compatibility is strongly related to the Flory-Huggins interaction parameter (χ) between donor and acceptor materials, which can be obtained by a simple differential scanning calorimetry measurement.[102] Experimental and computational data have demonstrated that an optimum phase purity and a high FF can be obtained only when χ is decently large to cause moderate phase separation in amorphous systems.[103] Some attempts have been made to tune the miscibility of donor and acceptor via molecular structure adjustment. For instance, to increase the χ of the hyper-miscibility system PB3T-C66:IT-4F, Duan and coworkers introduced the cyano group to the donor PB3T-C66, improving the domain purity from 0.62 to 1, and significantly increasing the JSC and FF.[105] Finally, the PCE was increased from 2.3% to 11.2%.[103] From the perspective of the acceptor, Hou and coworkers changed the end group of BTP-4Cl to reduce its miscibility with P3HT.[104] The enhanced χ of the system drives an appropriate phase separation, increasing the JSC and FF significantly.[104] In addition, the side chains of both donor and acceptor materials also affect the phase separation because the variation of solubility changes the χ parameter of the system.[105,106] However, there are still open questions about the relationship between molecular structure and miscibility. For example, how does the donor core of the NFA influence the compatibility of the system? What is the relationship between the polydispersity of the polymer donor and χ parameter of the system?[107] These are important questions that require further investigation.

It should be noted that high phase purity is often accompanied by large domain size, which is detrimental for charge separation. Recently, Ye et al. performed a detailed analysis of several typical PDCBT-Cl:nonfullerene systems from the viewpoint of mixing thermodynamics and film-solidification kinetics.[108] It was found that Y6 has excellent compatibility with PDCBT-Cl, and the blend of PDCBT-Cl:Y6 largely remains in the one-phase state. Hence, the device gave an extremely low JSC and FF, and thus a poor PCE of 0.53%. On the contrary, IDIC possesses high crystallinity and hypo-miscibility with PDCBT-Cl, leading to large phase separation. Therefore, PDCBT-Cl:IDIC also afforded a relatively low PCE of 9.44%. However, moderate miscibility between PDCBT-Cl and ITIC-Th1 helped the formation of high domain purity and suitable domain size simultaneously. It is close to the electron percolation threshold after proper post-treatment, thereby leading to the best efficiency of 12.11% with high JSC (18.3 mA cm⁻²) and FF (70%). Overall, an appropriate balance between phase purity and domain size is required for high efficiency.

Molecular Stacking: After exciton dissociation, what is important for the device performance is charge transfer and transport, which rely much on the molecular stacking and orientation. One of the structural features of FREAs is that the intermolecular interaction is formed via the π-π stacking of the terminal IC-group rather than the central core, due to the bulky group hanging from the molecular plane (Figure 5a–d). The reported crystalline data of ITIC-like NFAs shows that the terminal π-π stacking distance is about 3.5 Å (Figure 5d).[109–112] For example, IDT2Se-4F shows 2D interactions through terminal π-π stacking parallel to the ac plane and grid-like packing structures of conjugated backbones in the ab plane (Figure 5b,c).[112] This strong 2D transport network is beneficial for efficient electron transfer and transport. Moreover, this long-range structural ordering of non-fullerene along the backbone direction was also revealed by Grazing-Incidence Wide-Angle X-ray Scattering (GIWAXS) in the blend film, rationalizing the superior performance of FREA OSCs (Figure 5e,f).[113] It should be noted that the molecular stacking of FREAs is also affected by the polymer donor. Through quantitative analysis of the morphology of polymer:ITIC-Th blends and those state-of-the-art high-efficiency non-fullerene OSCs, a positive correlation between the (010) coherence length of the NFA in the active layer and the FF of the corresponding devices was found (Figure 5g). To some
extent, increasing the π-π stacking coherence length of NFAs via rationally designed polymer donors would be crucial for high-efficiency non-fullerene OSCs.\[114\]

Most reported NFAs provide 1D or 2D electron charge transport channel along the horizontal π-π stacking direction, limiting the electron mobility. Recently, some NFAs exhibit more than one set of transport channels (3D charge transport channel) to facilitate charge transport in all directions, similar to the isotropic transmission properties of fullerene acceptors, including both ITIC-like NFAs (e.g., ITIC-2Cl-δ) and Y-series NFAs (e.g., Y6, BTIC-CF3-γ).\[115–117\] Taking Y6 as an example, one Y6 molecule piles up on the top of two Y6 molecules through the end group stacking to form a twisted 1D transport channel in the single crystal (Figure 6a).\[116\] The banana-shaped molecules could extend their conjugation through end group stacking to form a zigzag polymer-like conjugated backbone. This special arrangement of molecules having two sets of transport channels facilitates the carrier transport; as a result, high electron mobility and current density were achieved. The single-crystal structure of BTIC-CF3-γ (a Y-series acceptor) reveals that the cooperated π-π interactions from H-aggregations of central fused cores and J-aggregations of end groups lead to a 3D interpenetrating network, which could be partially retained in the blend film (Figure 6b,c).\[117\] Thus it affords more charge transport channels with elevated electron mobility. As a result, the BTIC-CF3-γ-based solar cell achieved a high PCE of 15.59% with notable $J_{SC}$ (26.34 mA cm$^{-2}$) and FF (76.62%). Therefore, it is important to construct the 3D charge transport channels, which is determined by the pattern of NFA molecular stacking. However, how to control molecular stacking via NFA design remains unclear, calling for further attention.

Some other methods were also developed to tune the molecular stacking and phase separation in the active layer via molecular design, such as optimizing the conjugated length...
and backbone planarity via noncovalent interactions,\cite{118-120} and tuning the side-chain length to optimize the domain size and domain purity.\cite{24,106,121} They have been well documented elsewhere and will not be discussed in detail.

Post-Treatment Process: For specific donor/acceptor blends, various strategies of the post-treatment process for the morphology optimization have been reported, including the selection of processing solvent and additive, thermal annealing (TA), solvent annealing, and post solvent treatment for post-film morphology control.\cite{86,87,122-124} We will emphasize the post-treatment process, for example, the additive,\cite{56,123,125-127} ternary strategy,\cite{80,81,128-133} and interfacial modification\cite{132-135} here, which are the most used strategies in OSCs in recent years. The utilization of additives is a simple and effective method to change the film formation kinetics and promote molecular rearrangement.\cite{56,123,125-127} Various additives have been introduced to control the morphology of the active layer for high performance, including halogenous additives (e.g., 1,8-diiodooctane (DIO), chloronaphthalene (CN), etc.)\cite{136,137} and nonhalogenous additives (e.g., 1,8-octanedithiol (ODT), diphenyl ether (DPE), etc.).\cite{138,139} The additive was selected according to the two criteria of selective solubility and higher boiling point than the processing solvent.\cite{123} The solubility of NFAs in the additive plays a key role in controlling the morphology of the active layer, and hence the photovoltaic performance.\cite{56,86,123} Taking the FTAZ:IDCIC system as an example, the poor solubility of IDCIC in DIO promotes the IDCIC aggregation, enhancing the domain purity but enlarging the domain size; in contrast, its excellent solubility in CN inhibits the aggregation and facilitates IDCIC to diffuse into the donor, resulting in reduced domain size but decreased domain purity.\cite{56} As a result, the binary additive, CN&DIO, was employed to

Figure 6. a) Molecular packing sketch map of Y6 according to single crystal data. Reproduced with permission.\cite{116} Copyright 2020, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. b) Top view of the planar network structure of BTIC-CF3-γ. c) 3D interpenetrating network structure of BTIC-CF3-γ. Reproduced with permission.\cite{117} Copyright 2020, Elsevier Inc. The alkyl chains were ignored for clarity.
compromise the two effects. The FTAZ:IDCIC blend with CN&DIO demonstrated optimal domain size and high domain purity simultaneously, resulting in high device performance.

Apart from the traditional liquid additives, solid additives are an emerging approach to tune the morphology of the active layer.[27,140] For example, the solid additives with chemical structures similar to the end-groups of IT-4F, such as (E)-2-methyl-5-(thiophen-2-ylmethylene)-4H-cyclopenta[b]thiophene-4,6(5H)-dione (SA-1), enhance the intermolecular π–π stacking of the non-fullerene acceptor and thus facilitate the charge transport in the active layer of various NFA OSCs, leading to enhanced efficiencies.[140] The X-ray diffraction (XRD) and AFM images, shown in Figure 7a,b, suggest that SA-1 is well miscible with IT-4F. Moreover, the use of SA-1 enhances the intermolecular π–π interaction of IT-4F in both the annealed IT-4F+SA-1 film and the blend film. For the annealed PM6 film, the use of SA-1 has little influence on its crystallinity. According to the photovoltaic characterizations, XRD, and AFM results, the proposed working mechanism is shown in Figure 7c. During the spin-coating process, SA-1 may act as a small bridge to enhance the π–π stacking between two IT-4F molecules. Then SA-1 is removed from the film during the TA process, leaving more room for the self-assembly of IT-4F and forming a more condensed and ordered molecular arrangement, which makes it possible for a stronger π–π interaction among IT-4F molecules. As discussed in the section on phase separation, the donor matrix acts as a limit to the excessive movement of IT-4F molecules in this process, helping to maintain the good bi-continuous interpenetrating networks. As a result, the photovoltaic properties of the blend film were significantly improved by using SA-1 as the additive.

The ternary strategy is also one of the most effective methods for morphology control in non-fullerene OSCs.[122,130,131,141] Taking the advantages of both high charge transport of fullerene acceptors and outstanding light absorption of NFAs, the donor:NFA:fullerene acceptor system becomes the most popular and successful ternary solar cells in recent years.[61,122,130,142] For example, all small-molecule ternary solar cells with BTR:NITI:PC71BM showed a hierarchical morphology (Figure 8).[130] The third component, PC71BM, plays a critical role in separating the NITI and BTR phases and forming a morphological framework that provides efficient electron transport. The NITI and BTR form a smaller size phase separation and fit into the mesh of PC71BM network. Upon solvent vapor annealing (SVA), the BTR crystallization can push the NITI molecules out, leading to material

Figure 7. a) The corresponding XRD patterns and that of SA-1 film without TA. b) The corresponding AFM height images (A specified scale bar is used for the image of PBDB-TF+SA-1 film). c) Schematic diagram of the proposed working mechanism of solid additives. Reproduced with permission.[140] Copyright 2018, Springer Nature Limited.
enrichment at the PC71BM boundary. The PC71BM framework and good BTR crystallinity balance the carrier transport and reduce bimolecular recombination, contributing to high $J_{SC}$ and FF. In addition to PCBM, the amorphous bisadduct fullerene acceptor, ICBA (Figure S3, Supporting Information), has also been used to inhibit the π-π stacking of the crystalline non-fullerene acceptor ITIC-2Cl and helps achieve uniform morphology in the ternary blend. Therefore, it enables efficient charge dissociation, negligible bimolecular recombination, and balanced charge carrier mobilities.\(^{[122]}\)

The morphology of the active layer is also critically affected by the properties of the underlying interfacial layer. It has been reported that the surface free energy ($\gamma$) of the underlying interfacial layer can affect the molecular orientation in the active layer.\(^{[124,143]}\) For example, in PNTz4T:PC71BM blend film, PNTz4T is prone to adopt edge-on orientation when the substrate has low $\gamma$, leading to low mobility along the out-of-plane direction.\(^{[143]}\) Recently, tungsten oxide (WO$_x$) nanoparticles, poly(styrene sulfonic acid) sodium salts, and nickel formate dihydrate were introduced into poly(3,4-ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS) and successfully modified its $\gamma$. As a result, impressive FFs of approximately 80% have been achieved, which may be ascribed to the improved molecular orientation along the charge transport direction.\(^{[122]}\)

2.2. How to Maximize $V_{OC}$?

With the development of materials and device optimization in the past few years, all the parameters of OSCs, including $V_{OC}$, FF, and $J_{SC}$ have been improved greatly. When compared with the state-of-the-art high-efficiency solar cells (for example perovskite solar cells), although further improvement in the FF and $J_{SC}$ of OSCs can be expected, $V_{OC}$ might be the parameter that can bring about the next breakthrough of OSCs. To make an OSC efficient, $V_{OC}$ needs to be enhanced without sacrificing $J_{SC}$ and FF. Hence, it is necessary to decrease the energy loss for higher $V_{OC}$. For any kind of single-junction solar cell, the energy loss consists of three parts:

\[
q\Delta V = E_g - qV_{OC} = (E_g - qV_{OC}^{rad, below gap}) + (qV_{OC}^{rad, below gap} - qV_{OC}^{rad}) + (qV_{rad} - qV_{OC})
\]

where $q$ is the elementary charge, $\Delta V$ is the voltage loss, $E_g$ is the optical gap determined by the absorbing material with the narrowest gap in an OSC.\(^{[144]}\) $V_{OC}^{rad, below gap}$ is the maximum voltage according to the Shockley-Queisser limit, where the EQE is assumed to be 1 above the gap and 0 below the gap. $V_{OC}^{rad}$ is the open-circuit voltage when there is only radiative recombination, $V_{OC}^{rad, below gap}$ is the voltage loss of radiative recombination from the absorption below the gap, and $V_{OC}^{non-radi}$ is the voltage loss of the non-radiative recombination.

$\Delta E_1$, $(E_g - qV_{OC}^{rad})$ is due to radiative recombination originating from the absorption above the gap, which is unavoidable for any single-junction solar cell. As shown in Figure 9a, once the optical gap is fixed, $\Delta E_1$ is definite. For state-of-the-art OSCs, $\Delta E_1$ is typically around 0.25–0.30 eV.\(^{[145–147]}\)

$\Delta E_2$, $qV_{rad, below gap}$ is due to the additional radiative recombination from the absorption below the optical gap. According to the reciprocity relation,\(^{[148]}\) the radiative recombination is directly proportional to the photon absorption of the room-temperature blackbody radiation. As shown in Figure 9c, the room-temperature blackbody radiation at the low-energy region is much stronger than that in the high-energy region. As a result, the absorption below the optical gap can cause a large radiative loss. For inorganic or perovskite solar cells with...
steep absorption edges, $\Delta V_{\text{oc}, \text{rad}, \text{gap}}$ is negligible.\[^{33}\] For conventional OSCs based on the fullerene acceptors, the existence of CT states brings about the extra absorption within the optical gap (see Figure 9a,b). Despite the weak absorbance of these CT states due to their small oscillator strength with the ground state, those interfacial states generate a large amount of additional radiative recombination. For instance, the classical P3HT:PC$_{60}$BM OSCs show a $\Delta E_2$ as large as 0.67 eV.\[^{34}\] The situation in NFA-based blends is much different, with a wide range of blends demonstrating small $\Delta E_2$ values and efficient charge generation at the same time, including those based on ITIC (and derivatives), FBR (and derivatives), Y6 (and derivatives).\[^{35,149–153}\] In these cases, $\Delta E_2$ is mainly from the non-ideal absorption edge, as the CT state energy is close to the singlet exciton energy of low bandgap active materials, making the CT state absorption almost invisible.

$$\Delta E_2, \ (q\Delta V_{\text{oc}, \text{rad}} = -kT \ln(EQE_{\text{rad}})),$$

arises from the non-radiative recombination. $EQE_{\text{rad}}$ is the electroluminescence quantum efficiency of the solar cell when charge carriers are injected into the device in the dark. The enhanced $EQE_{\text{rad}}$ makes it possible to achieve low non-radiative energy losses. In other words, a good solar cell with low non-radiative recombination loss should also be a good light-emitting diode (LED). Since $\Delta E_2$ is unavoidable and $\Delta E_3$ is already small in state-of-the-art OSCs, $\Delta E_2$ plays a critically important role in the overall energy losses. Previous investigations on LEDs indicate that several conditions are required for high $EQE_{\text{rad}}$: 1) the number of electrons and holes injected should be as close as possible; 2) the probability of carriers leaving the diode without forming a bound electron-hole pair in bulk materials should be minimized; 3) the probability of generating photons following the recombination of electron-hole pairs should be maximized; 4) the photons generated in bulk materials should be efficiently outcoupled.

The emission properties of organic blends are largely determined by the intermolecular CT states at the interfaces between donor and acceptor materials. The luminescence of CT states can be influenced by the intrinsic molecular vibration, molecular packing, energetic disorder, HOMO or LUMO offsets between donors and acceptors, and so on. For example, an inherent link between non-radiative voltage losses and electron-vibration coupling has been found in fullerene-based OSCs, where the non-radiative recombination is enhanced for low-gap materials, following the energy-gap law.\[^{154}\] This was recently proved by an OSC device with a high $EQE_{\text{rad}}$ over 1% based on a wide-gap organic material blend due to the reduced electron-phonon coupling;\[^{155}\] however, the high $EQE_{\text{rad}}$ in this case was obtained by sacrificing the light absorption as wide-gap materials are used in this case, presenting a tradeoff between high $V_{\text{oc}}$ and high $J_{\text{sc}}$.

In contrast to fullerene-based systems, recent investigations on NFA OSCs indicate that the D/A blends with small LUMO or HOMO offsets show a low non-radiative loss, although the active materials in these cases show relatively narrow optical gaps.\[^{156}\] Quantum chemistry simulations demonstrate that CT states in blends with low energetic offsets can borrow the oscillator strength from the singlet excitons, which in the end enhances the radiative recombination of the blends. Within this framework, efficient radiative recombination can be achieved if the following conditions can be met: 1) the blend materials have a strong electronic coupling between CT and the first excited state, and 2) the narrow gap material has a high oscillator strength for transitions from the excited state to

Figure 9. a) The absorptance (blue curves) and emission (yellow curves) of Shockley–Queisser (SQ) type devices (top) and real-world OSC devices (bottom). In contrast to the absorptance of SQ type devices, the absorptance of real-world OSCs is not a step-function. Instead, the absorptance is smeared out with weakly absorbing sub-gap features attributed to the absorption of the CT state. These weakly absorbing features often dominate radiative recombination, which is the corresponding emission (yellow). Here, we note that, in some novel material systems, the interfacial CT state becomes nearly indistinguishable from the neat singlet excitons, leading to significantly reduced voltage losses. b) Energy level diagram depicting the energy of ground-state (S$_0$), local singlet (S$_1$) and triplet (T$_1$) excitons, CT state, local singlet (S$_1$) and triplet (T$_1$) excitons, CT state, and free carriers. The green arrow indicates optical absorption transitions within the neat narrow-gap material phase, and the yellow arrow indicates optical absorption by interfacial CT states. The red arrow indicates radiative excited-state decay. c) The photon flux density of 300 K blackbody radiation (only considering one-side absorption to mimic an OSC) and AM1.5G solar radiation (red) as a function of energy. The 300 K radiation is much higher at smaller energies than at high energies, which contributes greatly to the radiative recombination despite the weak absorption of the low-energy tail of the EQE. The product of solar spectra and EQE contribute to the charge generation and $J_{\text{sc}}$. 

$\Delta E_2 = -kT \ln(EQE_{\text{rad}})$
the ground state. The limitation is that small non-radiative voltage losses may come at the cost of higher overall recombination rates, which can explain the relatively low FF and EQE of some highly hybridized systems. 

Fortunately, this is not the case in the highest performing OSCs, where the small energetic offset does not seem to hinder free charge carrier generation. Sub-picosecond hole transfer has been observed in cases of moderate electronic coupling, wherein the high rates are attributed to a small reorganization energy. Additionally, electrostatic fields at the donor-acceptor interface have also been suggested to play a role in the charge separation of small offset cases. Recent theoretical quantum mechanical studies suggest that the non-radiative recombination depends on the molecular structures and their packing configuration; these calculations are consistent with experimental results where a face-on D/A interface undergoes less non-radiative recombination. Very recently, a low bandgap Y6 derivative, Y11, with low energetic disorder close to crystal silicon has achieved both high EQE and high PCE which might indicate the importance of the energetic disorder. In addition, Yip and coworkers combined experimental and theoretical modeling to find that the distinctive 3D molecular packing of Y6 leads to i) the formation of delocalized and emissive excitons that enable small non-radiative voltage loss, and ii) decalocalization of electron wavefunctions at D/A interfaces that significantly reduces the Coulomb attraction between interfacial electron-hole pairs. Thus it achieves highly efficient charge generation in PM6:Y6 systems with negligible donor-acceptor energy offset. In spite of these new developments, the thorough process of the dissociation and recombination in the systems with small energetic offsets is not fully understood. Furthermore, the relationship between electronic coupling, energetic disorder, reorganization energy, and electrostatic fields to effective material design needs further efforts as well.

In summary, the current methods to decrease non-radiative loss include increasing the gap of emissive species, tuning molecular packing, decreasing energetic disorder, and decreasing HOMO or LUMO offsets. Furthermore, an additional component, either donor/acceptor materials or neutral additives, can be added into the binary OSCs to reduce non-radiative recombination. As reported by Hou and coworkers, the non-radiative loss was decreased by 0.1 eV after adding PC61BM into PM6:Y6, leading to a PCE of over 16%. The suppressed non-radiative recombination can be possibly due to the fact that PC61BM can disperse the aggregation and suppress the self-quenching of Y6. A similar effect has been observed for a small molecular material NRM-1, which can effectively reduce non-radiative loss upon addition to some representative OSCs. We note that the addition of these extra components might also change other properties of the devices, for instance, the energetic disorder. As such, further investigations are still required to completely understand the role of PC61BM or NRM-1 and find more substitutes that are efficient.

3. Emerging Approaches in Enhancing the Device Stability in NFA OSCs

Stability is one of the key factors that limits the industrial process of OSCs. The inferior stability of OSCs results from the metastable morphology of the active layer, diffusion of the electrode and buffer, mechanical stress, oxygen and water, irradiation, heating, and so on. For more details, readers are suggested to consult those comprehensive reviews focusing on the stability of OSCs. Here we will discuss the influence of molecular structure, morphology, film processing, and device configuration on device stability.

3.1. NFA Molecular Structure

Organic semiconductors easily undergo photolytic and photochemical decomposition under illumination. The diffusion of oxygen molecules into the devices under ultraviolet (UV) light can generate superoxide radicals, which oxidizes organic semiconductors and results in irreversible photo-bleaching of the materials. Taking the classic ITIC as an example, upon different times of exposure to light in the air, the pristine film of ITIC fades gradually, whereas PC71BM retains most of the attenuation coefficient (Figure 10a). Compared with the PC71BM film, the absorption peak of ITIC significantly decreased in intensity and blue-shifted after exposure (Figure 10b). The molecular structure of ITIC contains more active photo-oxidation reaction sites than PC71BM, such as the double bond between the donor and acceptor units, the double bond on thiophene or bithiophene outlying central building blocks, side chains, etc. (Figure 10c). At these active reaction sites, the irreversible intercalation of oxygen atoms and the breakage of molecular backbones would be provoked by exposure to air. This will lead to increased trap states and increased energy loss, especially non-radiative recombination.

It has been reported that the stability of NFA molecules can be enhanced by the rational design of molecular structure. With the respect to the end groups, NFAs based on chlorine-substituted end groups exhibit improved photo-stability compared to those based on fluorine-substituted end groups. For instance, the optical density loss per hour (photo-bleaching rate) of IDIC-4Cl, IDIC-4F, and IDIC is ≈0.04%, ≈0.08%, and ≈0.4%, respectively. On the contrary, the methyl-substituted end groups showed decreased stability against light soaking. The ITIC-DM based devices suffer from strong burn-in loss of JSC and FF over the first several hundred hours of illumination, whereas the devices based ITIC-2F show promising long-term stability under the same conditions. The T80 (80% of the initial PCE) is estimated to be over 11 000 h for ITIC-2F based devices, which is quite encouraging for the development of NFAs. In terms of the donor moiety, it has been reported that applying non-fused donor cores is an effective strategy to improve the photostability of relevant NFAs. For example, non-fused electron acceptor PTIC based devices maintain about 70% of its initial PCE value for 50h illumination. However, the OSCs based on the fused counterpart ID-4F retain only 25% of its initial value under the same conditions.

3.2. Morphology

The device stability is related not only to the molecular structure, but also to the morphological stability. Molecular structures greatly influence their crystallinity and morphology
Figure 10. a) Photographs of photo-oxidized ITIC and PC₇₁BM pristine films upon different times of exposure. b) UV-visible spectra of photo-oxidized ITIC and PC₇₁BM pristine films upon different times of exposure. c) The primary photo-oxidation reaction sites of ITIC and PC₇₁BM. Reproduced with permission.[176] Copyright 2019, The Royal Society of Chemistry. d) Illustration of the $\chi$–$\Phi$ phase diagram for the hypomiscible and hypermiscible that corresponds to PTB7-Th:IEICO-4F and PTB7-Th:PC₇₁BM, respectively. Here $\Phi$ is the volume composition of the acceptor. $\Phi_i$ is the initial volume composition of the small molecule corresponding to the D/A weight ratio of 1:1.5. The percolation threshold is the composition of acceptor in the mixed region required to form a continuous electron transport pathway. Points F and A relate to freshly prepared and aged film, respectively. e) Schematic of morphology evolution corresponding to the burn-in degradation of PTB7-Th:IEICO-4F system and the stabilization of percolation via incorporation of PC₇₁BM. Reproduced with permission.[181] Copyright 2019, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.
properties, which are strongly associated with device stability. However, the relationship between the molecular structure-crystallinity-device stability has not yet been identified. Therefore, we will not discuss the morphology stability on the basis of molecular structure; instead, we discuss the morphology stability with respect to D/A miscibility and the ternary strategy.

From a thermodynamic perspective, excellent morphological stability is achieved when the miscibility ($\chi$) and component content ($\Phi$) are both optimum to reach the percolation threshold, which is an important standard for high efficiency. However, for most highly efficient blend systems, the percolation threshold difference between the two parameters ($\chi$, $\Phi$) is relatively large. In addition, solvents with different boiling points, solubility, and polarity also increase the difficulty of reaching the balanced percolation threshold between the miscibility and component content. For instance, the poor miscibility of ITIC/IDIC with PTB7-Th leads to morphological degradation during the photaging test, which accounts for the significant loss of $J_{sc}$ and FF. The improved miscibility of the D/A materials leads to a more stable morphology, which contributes to high efficiency and superior photostability in the PTB7-Th:EH-IDT devices. The $T_{50}$ of EH-IDT based devices was tested to be as long as 2132 h, which is much higher than that of ITIC and IDIC (only 221 and 598 h). The ternary strategy, which seems more general, is utilized to improve thermal stability and storage stability. How to choose the third component to stabilize the high-efficiency non-fullerene binary blend films through thermodynamics? Ade and coworkers proposed a morphological design rule to achieve stable and efficient non-fullerene OSCs: the ideal third component needs to have miscibility in the donor polymer at or above the percolation threshold, and be both thermodynamic and kinetic aspects. From the viewpoint of stability, the third component should have miscibility in the donor polymer at or above the percolation threshold, and be partly miscible with the crystallizable acceptor for stable morphology, as well as possess a high glass transition temperature for improved thermal stability.

In addition to lateral phase separation, vertical phase separation also affects the device stability significantly. As a result, the layer-by-layer (LbL, PHJ, and P-i-N) and pseudo-planar heterojunction structures show different stability, compared to the BHJ structure. In the J71:ITC6-IC blend film, for instance, the LbL film shows a dramatically different vertical phase separation compared with the BHJ blend, leading to improved device stability (Figure 11). From the results of time-of-light secondary ion mass spectrometry (TOF-SIMS), it was found that the BHJ blend has a donor-rich surface. For the LbL blend, the J71 donor was enriched at the bottom and the acceptor was assembled at the surface. The stability results indicate much suppressed degradation in the LbL system under illumination compared to the BHJ system (Figure 11d). The better light stability of LbL system was ascribed to the suitable vertical phase separation with enhanced donor and acceptor aggregations. The LbL device also shows better thermal stability due to its stable morphology (Figure 11e). Compared to the BHJ blend, the LbL morphology barely changed after baking at 120 °C for 1500 h. Furthermore, the LbL-bladed device showed enhanced bending stability, retaining 92% of its initial PCE after 2000 bending cycles with a radius of 6 mm (Figure 11f). Therefore, suitable vertical phase separation is an important factor for enhanced device stability. Further investigations about how to tune and control the vertical phase separation are needed.

### 3.3. Film Processing

Since blade-coating can induce a higher degree of molecular packing for both polymer donor and NFAs, it can partially replace the role of the additive DIO. That is, the blade-coating process realizes the optimized morphology and performance with fewer additives compared with the spin-coating method. For PBDB-T:ITIC, the blade-coated device with 0.25% DIO shows better stability than spin-coated devices with 1% DIO, due to the lower dissociation of residual DIO into iodotane and iodine radicals under illumination in the ambient environment as well as the slower morphology evolution with lower diffusion ability of donor and acceptor molecules. As a result, the blade-coated device with 0.25% DIO preserved 72.1% of the initial PCE, whereas the spin-coated device with 1% DIO only maintained 56.7% of the original PCE after 32 min of illumination.

### 3.4. Interfacial Contacts and Device Configuration

The interfacial contacts and device configuration also influence the stability, because the buffer layers and the electrodes exhibit mobility. For instance, the hole transport layer poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS) can diffuse into the active layer; the indium of indium tin oxide (ITO) can diffuse into PEDOT:PSS and the active layer. The diffusion of elements in the electrodes and the interlayers will reduce the stability of OSCs by changing the interfacial energy level alignments and forming traps for charge recombination. The interfaces and device configuration

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*Adv. Energy Mater. 2020, 10, 2002746* 2002746 (14 of 20) © 2020 The Authors. Advanced Energy Materials published by Wiley-VCH GmbH
for NFA OSCs need more attention because of the inferior stability of NFAs. In the PBDB-T:ITIC system, the NFA ITIC would react with PEDOT:PSS in a regular configuration, leading to a gradual decrease in the built-in potential and significant performance deterioration (58% decline in PCE during the aging test in air). This degradation can be suppressed by introducing a MoO₃ interfacial passivation layer. In addition, a continuous vertical phase separation process occurs during the aging period, leading to a PBDB-T-rich top surface and an ITIC-rich bottom surface in the active layer. As a result, the PBDB-T:ITIC based inverted devices, which are more favorable than the regular configuration ones for stable operation, only exhibit a 2.4% decrease in PCE during aging in air for 50 days.

4. Conclusion and Outlook

OSCs have developed significantly, owing to the advances in the fields of materials, devices, and mechanisms. However, there is still a long way to go before the widespread application is achievable. We now summarize these emerging approaches for enhancing the efficiency and stability of NFA OSCs, and highlight some aspects that might need further attention for further breakthroughs:

I. A wide range of narrow bandgap NFAs have been developed with great efforts to modify the donor moiety and electron-withdrawing end groups. These new NFAs, blended with donors having complementary absorption, are now increasingly closing the gap between OSCs and perovskite/inorganic solar cells, especially in terms of $J_{SC}$ and FF. In order to realize industrial applications, it is of critical importance to decrease the cost of NFAs, which can preferably be processed from green solvents and show good thickness tolerance. Along this line, more efforts should be dedicated to the simplification of the synthetic routes (for reduced cost), modification of the structures (for processing with green solvents), and improvement of the crystallinity of NFAs (for high mobility and hence thickness tolerance).

II. The energy losses have also been significantly decreased in NFA-based OSCs compared with those based on fullerene derivatives. In order to further decrease the energy losses to a level close to that of high-efficiency inorganic and perovskite solar cells, the key is to develop NFAs with strong photoluminescence, so that the non-radiative recombination losses can be suppressed. However, the relationship between the molecular structures and emission properties of NFAs remains unclear, and hence further efforts are needed to guide the design of highly emissive NFAs that maintain efficient charge generation at the same time. Meanwhile, it is quite interesting to note that some third components or additives can help to reduce non-radiative losses, although the underlying mechanism needs further clarification. It might be intriguing to have a deep understanding of the device physics in this case, so that we can also suppress the energy losses from the point of device engineering. In addition, the relationship between...
the morphology and energy losses, though discussed in some previous reports on fullerene-based OSCs, might also deserve further investigation in OSCs based on NFAs.

III. The stability of NFA OSCs is closely related to the organic materials and the morphology. The photo-oxidation of NFAs accounts for the degradation of devices, as they contain more active photo-oxidation reaction sites than the fullerene counterparts. It has been demonstrated that the photostability of NFAs can be improved by choosing the end groups and the donor moiety rationally. The structure-stability relationship requires more investigations so that the photostability of NFAs can be further enhanced. In terms of morphology stability, it is important to optimize the miscibility of D/A materials or introduce a third component to adjust the interaction of D/A materials. In addition, developing efficient and low-cost encapsulation is also very important for the commercial application of OSCs.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements
F.Z., H.Z., and R.Z. contributed equally to this work. F.Z. would like to thank the National Natural Science Foundation of China (Grant Nos. 21704021 and 21805288) and the European Commission Marie Skłodowska-Curie action (Grant No. 843872). F.G. would like to acknowledge the Stiftelsen for Strategisk Forskning through a Future Research Leader program (FFL18-0322), Swedish Research Council VR (Grant Nos. 2015-06146, 2018-05484, and 2018-06048), the Swedish Energy Agency (Grant No. 43691-1), and the Swedish Government Strategic Research Area in Materials Science on Functional Materials at Linköping University (Faculty Grant no. SFO-Mat-LiU #2009-00971).

Conflict of Interest
The authors declare no conflict of interest.

Keywords
device stability, narrow-gap, non-fullerene acceptors, voltage losses

Received: August 26, 2020
Revised: October 14, 2020
Published online: November 9, 2020

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current research interests include designing and fabricating low-energy-loss and high-performance solar cells.

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