Suppressing the excessive aggregation of nonfullerene acceptor in blade-coated active layer by using n-type polymer additive to achieve large-area printed organic solar cells with efficiency over 15%

Sheng Dong | Kai Zhang | Tao Jia | Wenkai Zhong | Xiaohui Wang | Fei Huang | Yong Cao

State Key Laboratory of Luminescent Materials and Devices, State Key Laboratory of Pulp & Paper Engineering, South China University of Technology, Guangzhou, China

Correspondence
Kai Zhang, Xiaohui Wang, and Fei Huang, State Key Laboratory of Luminescent Materials and Devices, State Key Laboratory of Pulp & Paper Engineering, South China University of Technology, Guangzhou 510640, China.
Email: mszhangk@scut.edu.cn (K. Z.);
Email: fewangxh@scut.edu.cn (X. W.);
Email: mshuang@scut.edu.cn (F. H.)

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Abstract
In this study, we demonstrate that high-efficiency blade-coated organic solar cells (OSCs) can be obtained by using polymer additive N2200 to suppress the excessive aggregation of nonfullerene acceptor. When using the blade-coating method, OSCs based on the blends of PM6:IT-4F and PM6:Y6 show moderate efficiencies of 9.9% and 13.2%, respectively. However, when a small amount of N2200 is added into the binary active layers, the resulting blade-coated OSCs show much higher efficiencies of 13.0% and 16.0%. Careful investigation reveals that the polymer additive suppresses the excessive aggregation of nonfullerene acceptors in the blade-coated active layer. This enhances the device performance by ensuring a more favorable morphology with appropriate domain size, improving charge extraction, and suppressing charge recombination. Based on this understanding, large-area OSC devices (1.0 cm²) are successfully obtained by blade coating, which display encouraging efficiencies of 12.3% and 15.1%.

KEYWORDS
blade coating, highly efficient, large-area device, organic solar cell, suppress aggregation

1 | INTRODUCTION

Bulk heterojunction (BHJ) organic solar cells (OSCs) have attracted extensive attention for their potential in fabricating large-area solar panels and flexible devices via low-cost solution-processing techniques.¹⁻⁵ In the past few years, OSCs have made great progress in terms of power conversion efficiency (PCE) owing to the rapid development of nonfullerene small-molecule acceptors, which possess excellent light absorption properties, high crystallinity, easy tunability of chemical structure, and less energy loss.⁶⁻¹⁵ Recently, the PCEs of single-junction OSCs over 16% have been achieved.
benefiting from the design of novel high-performance materials and the optimization of the active layer morphology and device engineering.\textsuperscript{16-20} However, most of these state-of-the-art OSCs are small-area devices and fabricated by spin coating in an inert atmosphere, which is poorly compatible with future massive production.\textsuperscript{21-23}

As a printing technique, blade coating is a widely used attractive alternative for realizing large-area OSCs because of its good compatibility with roll-to-roll processing.\textsuperscript{24-26} However, in recent studies, the optimal processing conditions for spin-coated devices have been proved inapplicable in achieving printed high-performance OSCs due to the different film-forming dynamic behaviors of spin coating and blade coating.\textsuperscript{27-29} During the spin-coating process, the films will solidify rapidly due to high spinning speed of the substrate and the strong centrifugal effect. However, in the case of blade coating, it will take a much longer time for the wet films to solidify. The slow solidification gives the active layer long time to aggregate and crystallize, which can lead to the formation of oversized domain or excessive phase separation in the active layer, especially for the currently reported high-performance nonfullerene acceptors with high crystallinity and strong aggregation tendency.\textsuperscript{29} Therefore, spin-coating and blade-coating techniques will often produce films with different nanoscale morphological features (phase separation, domain size), and result in a large variation in device performance. To achieve high-performance printed OSCs, some efforts have been attempted to optimize the nanoscale morphology of blade-coated active layer films, such as altering the processing solvents and additives, or adopting novel active layer materials.\textsuperscript{29-32} Although these strategies have succeeded to some extent in achieving efficient blade-coated devices, the gap between spin-coated and blade-coated device performance still remains. Moreover, scaling up a small-area device to a large-area device while retaining high performance remains a major challenge.

In this study, we demonstrate high-performance blade-coated OSCs by adding a small amount of n-type polymer N2200 in the active layer. The blade-coated OSCs based on PM6:IT-4F:5\% N2200 and PM6:Y6:10\% N2200 show encouraging PCEs of 13.0\% and 16.0\%, respectively, which are much higher than their counterpart devices without the polymer additive (9.9\% for PM6:IT-4F and 13.2\% for PM6:Y6). It has been found that, with the addition of N2200, the strong aggregation of nonfullerene acceptors during the blade-coating process is efficiently suppressed, leading to blend films with reasonably small domains, which in turn ensure efficient charge extraction and suppressed charge recombination. More in-depth studies reveal that N2200 prefers to suppress the aggregation of highly crystalline nonfullerene acceptors by incursion into the acceptor phase, benefiting from the better miscibility between them. On the basis of these results, efficient large-area OSCs are successfully designed and fabricated. With a device area of 1.0 cm$^2$, these blade-coated OSCs exhibit exciting PCEs of 12.3\% for PM6:IT-4F:5\% N2200 and 15.1\% for PM6:Y6:10\% N2200, showing an extremely low efficiency loss when the device area is scaling up.

## 2 RESULTS AND DISCUSSION

The chemical structures of PM6, IT-4F, Y6, N2200, and PNDIT-F$_3$N are shown in Figure 1A. The UV-vis absorption spectra of active layer materials are shown in Figure S1. The absorption onsets of IT-4F and Y6 are located at 793 nm and 931 nm, respectively. And the corresponding optical bandgaps estimated from the absorption onset are 1.56 eV for IT-4F and 1.32 eV for Y6. The absorption onset of N2200 is 831 nm corresponding to an estimated optical bandgap of 1.49 eV. The alcohol-soluble conjugated polymer PNDIT-F$_3$N was used as the cathode interlayer due to its good interface modification ability and high stability.\textsuperscript{33} Figure 1B shows a schematic energy-level diagram of the materials used in the study, and Figure S2 shows the related cyclic voltammetry (CV) curves. Figure 1C shows a diagrammatic drawing of the spin-coating and blade-coating processes. First, we took a blend of PM6 and IT-4F as the active layer to fabricate the OSCs based on spin coating with the device structure of indium tin oxide (ITO)/poly(3,4-ethylenedioxythiophene): polystyrenesulfonic acid (PEDOT:PSS)/PM6:IT-4F/4\%poly(9,9-bis’-(N,N-dimethylamino)propyl)-2,7-fluorene)-alt-5,5’-bis (2,2’-thiophene)-2,6-naphthalene-1,4,5,8-tetracaboxyl-N,N’-di(2-ethylhexyl)jimide)/Ag. The nonhalogenated solvent o-xylene (XY) was used as the processing solvent for active layer deposition, and various nonhalogenated additives were tested to optimize the morphology of the active layers to obtain high-performance OSCs. The corresponding photovoltaic characteristics of the spin-coated devices are summarized in Tables S1 and S2, and the relevant current density-voltage ($J$-$V$) curves are presented in Figures S3 and S4. The best-performing device was obtained with 0.5% dibenzyl ether (DBE) as the additive and provided a high PCE of 12.8\% with an open-circuit voltage ($V_{OC}$) of 0.85 V, a short-circuit current density ($J_{SC}$) of 20.6 mA cm$^{-2}$, and a fill factor (FF) of 73\%, which is comparable with other reported values.\textsuperscript{12} And then, we made printed OSCs by blade coating in the atmosphere by following the optimal solution formula used in the spin coating. The corresponding photovoltaic characteristics of the blade-coated devices are summarized in Table 1, and the relevant $J$-$V$ curves are...
As shown, when the coating method was changed from spin coating to blade coating, the best PCEs of the blade-coated devices dropped significantly to 9.9% along with a $V_{oc}$ of 0.82 V, a $J_{sc}$ of 18.9 mA cm$^{-2}$, and a FF of 64%. Figure 2C shows the external quantum efficiency (EQE) curves of the devices fabricated by spin coating and blade coating. As is shown, the EQE of the blade-coated device is obviously lower than that of the spin-coated device across the whole EQE response region.

Since the spin-coated and blade-coated devices had identical active layers and device structures, the difference in efficiency can presumably result from morphological effects. Atomic force microscopy (AFM) and transmission electron microscopy (TEM) were therefore employed to investigate the surface morphologies and phase separation of the active layer films deposited by using the two coating methods. As shown in Figure 3, the PM6:IT-4F film processed by spin coating shows a very smooth surface with a roughness less than 2 nm and an ideal bicontinuous networks (Figure 3A,E).

However, when deposited by blade coating, the morphology of the PM6:IT-4F film changed a lot. As shown in Figure 3B,F, the surface roughness of the blade-coated film increases to 10.83 nm, and large domains appear in the film, which were verified to arise from IT-4F aggregation (Figure S5, AFM images). Apparently, such strong aggregation would be detrimental to exciton dissociation and charge transport in the resulting devices, which is presumably responsible for the poor device performance.29

As demonstrated earlier, the blade-coated OSCs show poor photovoltaic performance, mainly attributed to the strong aggregation of the highly crystalline nonfullerene small-molecular acceptor IT-4F. Consequently, suppressing the excessive aggregation of IT-4F becomes quite crucial in achieving high-performance blade-coated devices. Here, we incorporated an n-type polymer additive N2200 into the blend of PM6:IT-4F to tune the crystallization dynamic process in the blade-coated OSCs. N2200 was chosen because it is a widely used n-type polymer, which can be expected not to affect the electric

**FIGURE 1**  A, Chemical structures of active layer materials and interlayer material used in the work; B, schematic energy-level diagram; C, illustration of spin-coating and blade-coating methods

| Active layer    | Coating method | $V_{oc}$ (V) | $J_{sc}$ (mA cm$^{-2}$) | FF (%) | PCE$_{avg}$ (%) | PCE$_{max}$ (%) |
|-----------------|----------------|-------------|-------------------------|--------|----------------|----------------|
| PM6:IT-4F       | Spin coating   | 0.85        | 20.6                    | 73     | 12.5           | 12.8           |
| PM6:IT-4F       | Blade coating  | 0.82        | 18.9                    | 64     | 9.2            | 9.9            |
| PM6:IT-4F:N2200 | Spin coating   | 0.86        | 21.1                    | 74     | 13.3           | 13.5           |
| PM6:IT-4F:N2200 | Blade coating  | 0.86        | 20.8                    | 73     | 12.8           | 13.0           |
Moreover, N2200 is also used as the third component in ternary OSCs, in which N2200 can enhance electron transport and improve device photostability of the resulting OSCs.\textsuperscript{36} To evaluate the effect of the N2200 additive on the photovoltaic performances of PM6:IT-4F devices, OSCs were firstly fabricated by spin coating with various weight ratios of N2200. The corresponding photovoltaic characteristics of

\textbf{FIGURE 2} A,C, \textit{J–V} and external quantum efficiency (EQE) curves of the spin-coated and blade-coated devices based on the blend of PM6:IT-4F. B,D, \textit{J–V} and EQE curves of the spin-coated and blade-coated devices based on the blend of PM6:IT-4F:N2200

\textbf{FIGURE 3} A–D, Atomic force microscopy (AFM) images of the spin-coated and blade-coated active layer films based on the blend of PM6:IT-4F and PM6:IT-4F:N2200. E–H, TEM images of the spin-coated and blade-coated active layer films based on the blend of PM6:IT-4F and PM6:IT-4F:N2200. I, X-ray diffraction (XRD) data of pure and blend films fabricated by blade coating
the spin-coated devices are summarized in Table S3, and the relevant J-V curves are presented in Figure S6. The J-V curve of the best-performing device is shown in Figure 2B, and the corresponding device data are listed in Table 1. The best-performing device was obtained with 5 wt% N2200 and provided a high PCE of 13.5%, slightly higher than the binary device, with a $V_{oc}$ of 0.86 V, a $J_{sc}$ of 21.1 mA cm$^{-2}$, and a FF of 74%. And then, the blade-coated devices based on the active layer of PM6:IT-4F: N2200 were fabricated. Encouragingly, the blade-coated devices with N2200 showed a significantly enhanced photovoltaic performance, where the best device had a PCE of 13.0%, a $V_{oc}$ of 0.86 V, a $J_{sc}$ of 20.8 mA cm$^{-2}$, and a FF of 73%, outperforming the blade-coated binary device. More importantly, the tiny difference in PCE between the spin-coated and blade-coated ternary devices (13.5% vs 13.0%) demonstrates that the device efficiency can be retained when changing the coating method from spin coating to blade coating. Moreover, the EQE of the blade-coated device with N2200 is comparable to that of the spin-coated device in the whole EQE response region (Figure 2D).

To explore the relationship between the improved photovoltaic performance and the nanoscale morphology, we further used AFM and TEM to investigate the N2200-containing blade-coated active layer. As shown in Figure 3D,H, in comparison with the high surface roughness and large domain size of the blade-coated PM6:IT-4F film, the PM6:IT-4F:N2200 film processed by blade coating shows a much smoother surface with a roughness of 2.12 nm and small domains. The surface morphology and phase separation of this film are quite similar to those of the spin-coated films (Figure 3C,G). To further characterize the effect of N2200 on the morphology of the blade-coated active layer, we carried out X-ray diffraction (XRD) test. As shown in Figure 3I, a clear diffraction peak around 5.8° is observed in the IT-4F and PM6:IT-4F films, which is ascribed to the crystallization of IT-4F. Meanwhile, the diffraction peak around 4.4° can be ascribed to PM6 in the PM6:IT-4F films. However, in the PM6:IT-4F:N2200 film, the diffraction intensity of IT-4F is largely suppressed but a strong diffraction of PM6 still exists, indicating that the crystallization of IT-4F is hindered due to the existence of N2200, and in turn efficiently suppressing the strong aggregation trend. That is to say, although IT-4F in the binary blends tended to form large aggregates during blade coating due to its high crystallinity, N2200 suppressed this aggregation and thus directed the morphological evolution of the active layer toward a more favorable structure. Considering that N2200 selectively suppressed the aggregation of IT-4F, we speculated that the hindered crystallization may be due to the incursion of N2200 into IT-4F. To verify this hypothesis, the surface energies of the pure films were estimated from the contact angles of pure water and diiodomethane. As shown in Figure S9 and Table S4, the surface energy of N2200 was estimated to be 42.3 mN m$^{-1}$, which is closer to that of IT-4F (46.4 mN m$^{-1}$) than that of PM6 (33.4 mN m$^{-1}$), indicating that N2200 and IT-4F had better miscibility with each other than with PM6, and would tend to form a mixture in the blend film. This result suggests that N2200 molecules would tend to localize in IT-4F domains and thus suppress their aggregation.

A potential concern is that the charge mobility of the PM6:IT-4F film would be reduced due to the incursion of N2200. To test this, the hole-only and electron-only devices based on binary and ternary films were prepared, and the hole and electron mobilities were extracted by fitting the data using an space-charge-limited current model. As shown in Figure S10 and summarized in Table S5, for the spin-coated film of PM6:IT-4F, the electron and hole mobilities ($\mu_e$ and $\mu_h$) are $4.39 \times 10^{-4}$ and $1.14 \times 10^{-3}$ cm$^2$ V$^{-1}$ s$^{-1}$, respectively. However, for the blade-coated film of PM6:IT-4F, $\mu_e$ and $\mu_h$ decrease to $2.23 \times 10^{-4}$ and $8.04 \times 10^{-4}$ cm$^2$ V$^{-1}$ s$^{-1}$, respectively. The most likely cause of this reduced mobility is the large size of the IT-4F domains, which acts as an obstacle to charge hopping between adjacent phases. To our delight, for the blade-coated film of PM6:IT-4F:N2200, $\mu_e$ and $\mu_h$ are improved to $4.31 \times 10^{-4}$ and $1.12 \times 10^{-3}$ cm$^2$ V$^{-1}$ s$^{-1}$, which are comparable to the spin-coated binary film, demonstrating that N2200 does not disturb the charge transport. On the contrary, the incorporation of N2200 leads to the buildup of efficient charge transport pathways by restricting the phase separation to acceptably small domains. Correspondingly, the $\mu_h$/ $\mu_e$ ratios of the blade-coated PM6:IT-4F and PM6:IT-4F: N2200 films are 3.61 and 2.60, respectively. Both the overall enhancement and the improved balance of the charge mobilities contributed to reducing the accumulation of space charges and improving the $J_{sc}$ and FF values.

Grazing-incidence wide-angle X-ray scattering (GIWAXS) was also measured to investigate the structure order and textures of the active layer films. Figure 4 illustrates the two-dimensional (2D) patterns and the corresponding out-of-plane and in-plane line-cuts. For the PM6:IT-4F film fabricated by spin coating (Figure 4A), a preferential face-on orientation with respect to the substrate is evidenced by the distinct (010) diffraction peak of strong $\pi-\pi$ stacking in the out-of-plane direction, which is favorable for the vertical charge transport. However, for the PM6:IT-4F film fabricated by blade coating (Figure 4B), the (010) diffraction peak at $q \sim 1.7$ Å$^{-1}$ becomes indistinct and face-on orientation is weaken. Interestingly, when a small amount of N2200 was added to the PM6:IT-4F film, the blade-coated film (Figure 4D) displays a more pronounced diffraction peak in the out-of-plane direction located at $q \sim 1.7$ Å$^{-1}$, which indicates the strengthening of the face-on oriented $\pi-\pi$ stacking. Moreover, compared with the spin-coated ternary films
Figure 4  A-D, GIWAXS 2D patterns and (E) corresponding out-of-plane (solid lines) and in-plane (dashed lines) line-cut profiles of the active layer films based on the blend of PM6:IT-4F and PM6:IT-4F:N2200 processed by different coating methods.

Figure 5  A, $J_{sc}$; B, $V_{oc}$ as a function of light intensity; C, transient photocurrent measurements of the blade-coated organic solar cells (OSCs) based on the blend of PM6:IT-4F and PM6:IT-4F:N2200. D, $J$–$V$ curve of the large-area blade-coated device based on PM6:IT-4F:N2200 and PM6:Y6:N2200, with an inset of photographs of the 1.0 cm² blade-coated device.
Figure 5B shows the N2200 efficiently retarded bimolecular recombination. PM6:IT-4F (0.942), which indicates that the addition of higher than that for the blade-coated device based on blade-coated device with PM6:IT-4F:N2200 (0.987) is affected by the overall device performance, we studied the dynamics of charge recombination and charge extraction. The primary charge recombination behaviors in blade-coated devices based on PM6:IT-4F and PM6:IT-4F: N2200 were investigated by measuring the dependence of the $J-V$ characteristics on light intensity ($P_{\text{light}}$). As shown in Figure 5A, the $a$ value of the fitted line for the blade-coated device with PM6:IT-4F:N2200 (0.987) is higher than that for the blade-coated device based on PM6:IT-4F (0.942), which indicates that the addition of N2200 efficiently retarded bimolecular recombination. Figure 5B shows the $V_{\text{oc}}$ as a function of $P_{\text{light}}$ The slope for the blade-coated device with PM6:IT-4F:N2200 is 1.16 $kT/q$, which is lower than that of the blade-coated device with PM6:IT-4F (1.78 $kT/q$), indicating that Shockley-Read-Hall recombination was suppressed due to the addition of N2200. Next, transient photocurrent measurements were taken to study the competition between charge recombination and charge extraction in the OSCs. As shown in Figure 5C, the charge extraction time is 0.28 μs for the blade-coated device with binary PM6:IT-4F, which decreases to 0.16 μs for the device with the N2200 additive. The much shorter charge extraction time suggests more efficient electron extraction, which leads to enhanced device performance.

The above results suggest a strategy to obtain high-performance blade-coated OSCs. To test the universality of this strategy, OSCs based on PM6:Y6 as active layers were studied. The corresponding photovoltaic characteristics of the devices made by the two coating methods are summarized in Table 2 and Table S6, and the relevant $J-V$ curves are presented in Figures S11 and S12. As is shown, the spin-coated OSCs based on the blend of PM6:Y6 shows a high PCE of 15.8%, which is comparable with the previously reported value. However, when the coating method was changed from spin coating to blade coating, the PCE of the PM6:Y6-based device significantly dropped to 13.2%, which is mainly attributed to the aggregation of Y6 during the blade-coating process (Figure S13). When adding 10 wt% N2200 into the blend of the binary active layer, the resulting spin-coated and blade-coated OSCs show significantly enhanced photovoltaic performance with much higher PCEs of 16.6% and 16.0%, respectively. The EQE curves of binary and ternary devices based on PM6:Y6 and PM6:Y6:N2200 are shown in Figure S14. As is shown, the $J_{\text{sc}}$ values calculated from the EQE curves are comparable to those measured from the $J-V$ curve with mismatch less than 5%. This result demonstrates that suppressing the strong aggregation of active layer during blade coating is a general method to achieve efficient blade-coated OSCs.

Finally, in order to evaluate the applicability of this strategy to large-area OSCs, we further fabricated large-area devices (1 cm$^2$) by using blade coating in a fume hood. As shown in Figure 5D and Table S7, the large-area devices exhibit encouraging PCEs of 12.3% for PM6:IT-4F:N2200 and 15.1% for PM6:Y6:N2200. To the best of our knowledge, our device should be the first reported blade-coated large-area OSCs with PCE over 15% until now.

### 3 | CONCLUSION

In summary, we demonstrated an effective strategy to achieve high-performance blade-coated OSCs by simply introducing an n-type polymer additive N2200 to the active layer blend of PM6:IT-4F or PM6:Y6. The blade-coated OSCs based on PM6:IT-4F:N2200 and PM6:Y6: N2200 showed encouraging PCEs of 13% and 16%, much higher than their counterpart devices without N2200 (9.9% for PM6:IT-4F and 13.2% for PM6:Y6). Careful research studies showed that N2200 prefers to prevent the evolution of nonfullerene acceptors into excessively large-size domains in the blade-coated film by incursion into the acceptor phase, benefitting from the better miscibility between them, and thus improved the device performance. Furthermore, we fabricated large-area devices (1.0 cm$^2$) by using blade coating and obtained highly encouraging PCEs of 12.3% for PM6:IT-4F:N2200 and 15.1% for PM6:Y6:N2200. These results indicated that suppressing the strong aggregation of the active layer

| Active layer | Coating method | $V_{\text{oc}}$ (V) | $J_{\text{sc}}$ (mA cm$^{-2}$) | FF (%) | PCE$_{\text{avg}}$ (%) | PCE$_{\text{max}}$ (%) |
|--------------|----------------|---------------------|---------------------|--------|---------------------|---------------------|
| PM6:Y6       | Spin coating   | 0.82                | 25.7                | 74     | 15.5                | 15.8                |
| PM6:Y6       | Blade coating  | 0.81                | 24.4                | 66     | 12.6                | 13.2                |
| PM6:Y6:N2200 | Spin coating   | 0.83                | 26.3                | 76     | 16.3                | 16.6                |
| PM6:Y6:N2200 | Blade coating  | 0.83                | 26.3                | 74     | 15.6                | 16.0                |

(Figure 4C), the blade-coated ternary films exhibit slightly stronger and more pronounced scattering peaks, indicating that blade coating induced a higher degree of molecular packing.

To further understand how the addition of N2200 affected the overall device performance, we studied the dynamics of charge recombination and charge extraction. The primary charge recombination behaviors in blade-coated devices based on PM6:IT-4F and PM6:IT-4F: N2200 were investigated by measuring the dependence of the $J-V$ characteristics on light intensity ($P_{\text{light}}$). As shown in Figure 5A, the $a$ value of the fitted line for the blade-coated device with PM6:IT-4F:N2200 (0.987) is higher than that for the blade-coated device based on PM6:IT-4F (0.942), which indicates that the addition of N2200 efficiently retarded bimolecular recombination. Figure 5B shows the $V_{\text{oc}}$ as a function of $P_{\text{light}}$. The slope for the blade-coated device with PM6:IT-4F:N2200 is 1.16 $kT/q$, which is lower than that of the blade-coated device with PM6:IT-4F (1.78 $kT/q$), indicating that Shockley-Read-Hall recombination was suppressed due to the addition of N2200. Next, transient photocurrent measurements were taken to study the competition between charge recombination and charge extraction in the OSCs. As shown in Figure 5C, the charge extraction time is 0.28 μs for the blade-coated device with binary PM6:IT-4F, which decreases to 0.16 μs for the device with the N2200 additive. The much shorter charge extraction time suggests more efficient electron extraction, which leads to enhanced device performance.

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during blade coating with a polymer additive is a promising strategy to achieve efficient large-area printed OSCs, and we also demonstrated the potential application of this strategy in massive production.

4 EXPERIMENTAL SECTION

4.1 The fabrication of OSCs

ITO-coated glass substrates were sequentially cleaned with detergent, acetone, deionized water, and isopropanol, and then dried at 60°C in a baking oven overnight. After 3 minutes of plasma treatment, the precleaned ITO substrates were coated with 40 nm PEDOT:PSS (Clevios P VP AI 4083) by spin coating and then baked at 140°C for 15 minutes. PM6 and IT-4F (1:1, w/w) were dissolved in o-xylene with DBE as the additive, and PM6 and Y6 (1:1.2, w/w) were dissolved in chloroform with 0.5% chloronaphthalene overnight on a hot plate at 50°C. For spin-coated devices, active layers were formed by spin coating these solutions onto substrates. For the blade-coated device, active layer solutions (15 μL) were coated on top of PEDOT layer using a doctor-blade coater (Erichsen, Berlin, Germany) in air at a coating speed of 10 mm s⁻¹, and the gap height was set to be 300 μm. After that, the active layer films were thermally annealed at 100°C for 10 minutes. Next, 10-nm cathode interlayer PNDT-F3N was spin coated onto the active layers. Finally, 100 nm Ag was evaporated through a shadow mask in a vacuum chamber at a base pressure of 1 × 10⁻⁷ mbar, with an active area of 0.0516 cm².

4.2 Device characterization

The J-V curves of all devices with a metal mask of 0.04 cm² to define the device area were measured on a computer-controlled Keithley 2400 SourceMeter under 1 sun, AM 1.5 G spectra from solar simulator (Enlitech, Kaohsiung, Taiwan). The light intensity was 100 mW cm⁻² as calibrated by using a China General Certification Centre (CGC) certified reference monocrystalline silicon cell (Enlitech). The EQE spectra were recorded using a QE-R measurement system (Enlitech, QE-R3011, Taiwan).

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CONFLICT OF INTEREST

The authors declare no potential conflict of interest.

ORCID

Fei Huang https://orcid.org/0000-0001-9665-6642

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SUPPORTING INFORMATION
Additional supporting information may be found online in the Supporting Information section at the end of this article.

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