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Efficient Organic Solar Cells Fabricated with a High-Boiling Solvent Via Morphology Modulation Using a Small Molecule Guest Donor

Longyu Li, Yuxin Wang, Zhaochen Suo, Wanying Feng, Wenkai Zhao, Guanghui Li, Zhaoyang Yao, Guankui Long, Xiangjian Wan,* and Yongsheng Chen

It is necessary to employ high-boiling solvents for the fabrication of large-area organic solar cells (OSCs). However, controlling the morphology to achieve suitable phase separation for high efficiency is challenging when high-boiling solvents are used. In this study, HD-1, a small molecular donor is introduced, into the PM6:BO-4Cl blend as a morphology-modulating agent. This addition optimizes the phase separation and enhances charge transport in the ternary blend processed in chlorobenzene (CB) solvent. The optimized active layer demonstrates improved charge dissociation and transport capabilities while effectively suppressing charge recombination. Small-area OSCs processed from the CB solution achieves an improved power conversion efficiency (PCE), increasing from 18.84% to 19.42%. Furthermore, the device demonstrated good tolerance of active layer thickness, with a 300-nm-thick film achieving a PCE exceeding 17%. Notably, a blade-coated module with an active area of 13.5 cm², fabricated in air using CB, achieves a PCE of 16.48%, showing excellent application potential. This study presents a straightforward and effective approach to optimizing active layer morphology and enhancing the PCE of OSCs fabricate using a high-boiling solvent, highlighting its potential for large-scale production and practical applications.

1. Introduction

Organic solar cells (OSCs) are highly regarded for their advantages including lightweight,^[1–3] flexibility,^[4–6] and compatibility

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with roll-to-roll processing,^[7-9] making them particularly well-suited for specific applications such as smart wearable devices,^[10–12] the Internet of Things (IoT),^[13] and building-integrated photovoltaics (BIPVs).^[14–16] In recent years, advancements in material systems and optimizations of active layer morphology have led to a steady increase in the power conversion efficiencies of OSCs,^[17-27] with efficiencies now exceeding 20%.[28-30] Due to the high exciton binding energy of organic materials, nanoscale fibrillar morphology with abundant interfaces is crucial for effective exciton dissociation and transport,[31-33] which are key to achieving high-efficiency OSCs. Currently, most high-efficiency devices, typically under 0.1 cm² in area, are fabricated using the spin-coating process with chloroform (CF) as the solvent.^[34-36] The fast evaporation kinetics of CF facilitate the formation of nanoscale phase-separation morphology during the film formation process.[37,38] However, significant material loss occurs

during the spin-coating process, and the volatile nature of CF makes it unsuitable for large-area production.^[39,40] Although some studies have demonstrated the potential for using CF in large-area OSCs, strict control of ambient temperatures is required,^[41,42] adding complexity to the process. High-boiling solvents, such as chlorobenzene (CB) and o-xylene provide a wide processing window for large-area device production. However, the use of these solvents can result in the excessive selfaggregation of active layer molecules during the film-forming process,^[43-46] potentially leading to excessive phase separation and significantly reduced PCE. Furthermore, for large-area module manufacture, particularly roll-to-roll processing, the active layer materials must exhibit thickness tolerance.^[47,48] Therefore, the development of high-efficiency, thickness-tolerant active layer material systems using high-boiling-point solvents is essential for large-area OSCs production.

In this study, we introduce HD-1,^[49] a small molecule donor highly miscible with BO-4Cl, into the PM6:BO-4Cl blend film. The introduction of HD-1 not only mitigates the over-aggregation of acceptors in the high-boiling-point solvent CB but also enhances and balances carrier mobility in the ternary blend film.

L. Li, Y. Wang, Z. Suo, W. Feng, G. Li, Z. Yao, X. Wan, Y. Chen State Key Laboratory of Elemento-Organic Chemistry Frontiers Science Center for New Organic Matter Key Laboratory of Functional Polymer Materials Institute of Polymer Chemistry Tianjin Key Laboratory of functional polymer materials Nankai University Tianjin 300071, China E-mail: xjwan@nankai.edu.cn W. Zhao, G. Long School of Materials Science and Engineering National Institute for Advanced Materials Nankai University Tianjin 300071, China





Figure 1. a) Chemical structures of PM6, HD-1, and BO-4Cl. b) Normalized film absorption spectra of PM6, HD-1, and BO-4Cl. c) Energy level diagram of the corresponding materials. d) Flory-Huggins interaction parameter χ of PM6:BO-4Cl, PM6:HD-1, and HD-1:BO-4Cl. e) PCE and FF of devices with different HD-1 ratios. f) *J*–V curves and g) EQE curves for PM6:BO-4Cl and PM6:HD-1:BO-4Cl solar cells.

As a result, after incorporating HD-1 as a guest donor into the binary device, an impressive efficiency of 19.42% was achieved. Furthermore, the 300-nm-thick film device achieved a PCE exceeding 17%, and when scaled up, the large-area module prepared by blade coating in air reached a PCE of 16.48%. Our work demonstrates that using a small molecule donor with high charge mobility as the third component, which exhibits good miscibility with the host acceptor, offers a promising approach for fabricating efficient thick-film devices and modules.

2. Results and Discussion

The chemical structures of the photoactive materials, UV–vis absorption spectra of the pure films, and the energy level diagram are presented in **Figure 1a–c**. The complementary UV–vis absorption spectra of pure films effectively cover the visible to nearinfrared region, ensuring sufficient photon capture. Additionally, the cascaded energy levels of active layer materials facilitate efficient charge transport and collection in ternary devices.^[50] To quantitatively study the compatibility of HD-1 with host materials, the Flory-Huggins interaction parameter (χ) was calculated according to the detailed contact angle results of water and ethylene glycol on PM6, HD-1, and BO-4Cl neat films.^[51] The χ values of PM6:BO-4Cl, PM6:HD-1, and HD-1:BO-4Cl were determined as 1.45, 0.49, and 0.25 K, respectively (Figure 1d), indicating similar miscibility parameters for PM6 versus HD-1 and HD-1 versus BO-4Cl. This favorable miscibility supports the formation of good morphology and phase separation in the active layers.^[52-54]

To investigate the effects of introducing HD-1 into the PM6:BO-4Cl system, binary and ternary devices with varying HD-1 ratios were fabricated using a conventional device structure of indium tin oxide (ITO)/2PACz/PM6:HD-1:BO-4Cl/PNDIT-F3N/Ag. As shown in Figure 1e, the optimal HD-1 ratio for ternary devices was found to be 0.9:0.1:1.2 (PM6:HD-1:BO-4Cl). The best ternary device, with an active layer thickness of

Table 1. Photovoltaic performance of the devices.

	V _{oc} [V]	$J_{\rm SC}$ [mA cm ⁻²]	$\int_{CAL}^{a)}$ [mA cm ⁻²]	FF [%]	PCE ^{b)} [%]
PM6:BO-4Cl	0.854	28.08	27.20	78.40	18.84 (18.66 ± 0.16)
PM6:HD-1:BO-4Cl	0.848	28.56	27.60	80.00	19.42 (19.15 ± 0.17)
Module	5.043	4.44	-	73.57	16.48 (16.11 ± 0.20)

^{a)} J_{CAL} values are calculated from EQE curves; ^{b)} PCEs are average values with standard deviations calculated from eight individual devices.

≈130 nm, exhibited a PCE of 19.42%, a short-circuit current density (J_{SC}) of 28.56 mA cm⁻², and an FF of 80.00%. These improvements can be attributed to the optimized morphology, enabling efficient charge generation and transport. In contrast, control devices using PM6:BO-4Cl with a similar active layer thickness showed a lower I_{SC} and FF, resulting in a PCE of 18.84% (Figure 1f). External quantum efficiency (EQE) spectra for both binary and ternary devices, shown in Figure 1g, revealed that the calculated integrated J_{SC} values from the EQE spectrum closely matched the current densities derived from J-V curves (Table 1). The enhanced EQE and FF values in the ternary devices can be ascribed to the incorporation of HD-1, which optimizes the blend film morphology, improves charge carrier collection, and suppresses recombination. Increasing the active layer thickness to \approx 300 nm further demonstrated the superior performance of ternary devices. As shown in Figure S2 and Table S4 (Supporting Information), the 300 nm device achieved a promising PCE of 17.49%, with a $V_{\rm OC}$ of 0.83 V and an FF exceeding 70%. In contrast, the binary counterparts delivered a PCE of 16.51% with a lower FF of 67.50%, primarily due to increased charge recombination. Beyond photovoltaic performance, the stability of OSCs is equally important. Thanks to the optimized phase separation morphology of the active layer, ternary devices demonstrated enhanced thermal stability compared to binary devices. After being heated at 65 °C for 300 h, the PM6:BO-4Cl device retained 75.4% of its initial PCE, while the PM6:HD-1:BO-4Cl device retained 78.8% (Figure S3, Supporting Information). Additionally, the photostability of the devices followed a similar improvement trend.

To compare the charge recombination mechanisms in the devices, the dependence of $J_{\rm SC}$ and $V_{\rm OC}$ on varying light intensity (P_{light}) was measured. Typically, the relationship between J_{SC} and P_{light} follows the power law $J_{\text{SC}} \propto (P_{\text{light}})^{\alpha}$, where α represents the slope of the curve. Fitting the data in Figure 2a, α values of 0.991 and 0.995 were obtained for the 130 nm binary and ternary devices, respectively, indicating reduced bimolecular recombination in these devices. However, when the active layer thickness increased to \approx 300 nm, slightly lower α values were observed (Figure S4, Supporting Information). This can be attributed to a higher likelihood of recombination, as the extended time required for charge transfer to the electrodes in thicker films increases the chance of recombination. Additionally, the dependence of $V_{\rm OC}$ on Plight was characterized to further examine charge recombination behavior (Figure 2b). According to the relation $V_{\rm OC} \propto S(P_{\rm light})$ the S value for the ternary device (1.05 KT/q) was significantly lower than that of the binary device (1.11 KT q^{-1}), suggesting reduced trap-assisted recombination in the ternary devices. To gain deeper insights into light absorption and exciton dissociation processes, saturation current density (I_{sat}) measurements

and charge dissociation probability P (E,T) analyses were conducted for both binary and ternary OSCs.[55] The charge dissociation probability P (E,T) improved in the ternary devices, increasing from 97.6% to 98.9% under short-circuit conditions and from 89.7% to 90.4% under maximal power output conditions, compared to the binary device (Figure 2c). These results align with the trends observed in the EQE curves, further validating the performance enhancements in the ternary device. Transient photovoltage (TPV) and transient photocurrent (TPC) measurements were conducted to investigate the influence of reduced recombination behavior, as shown in Figure 2d,e. Upon incorporating HD-1, the device based on PM6:HD-1:BO-4Cl exhibited a faster charge extraction time of 0.34 us, shorter than the 0.45 us observed for the binary device. Additionally, the carrier lifetimes (τ) derived from TPV measurements were 52.9 µs for the binary device and 70.7 μ s for the ternary device. The longer τ observed in the ternary device indicates reduced charge recombination.^[56] These findings demonstrate that the inclusion of HD-1 improved charge transport and extraction while suppressing charge recombination, ultimately enhancing the J_{SC} and FF of the ternary device. Moreover, hole mobility $(\mu_{\rm h})$ and electron mobility $(\mu_{\rm e})$ were extracted using the space charge-limited current (SCLC) method. The ternary device exhibited higher hole/electron mobilities and a more balanced μ_h / μ_e ratio compared to the binary device (Figure 2f) indicating improved charge transport properties.^[57] These findings are consistent with the significantly improved FF and I_{SC} observed in the devices incorporating HD-1. The transient absorption (TA) spectrum was utilized to further study the charge-transfer behavior in blend films. The pump light at 800 nm was applied to excite the acceptor and investigate the process of hole transfer. The TA spectra of blend films are shown in Figure 2g,h. The negative bleaching signals at \approx 631 nm were assigned to the ground state bleach (GSB) of donors. The GSB signal of BO-4Cl decreased in sync with the increase of the GSB signals of the donors, which verifies the process of hole transfer from acceptor to donor. Then, the GSB kinetics were fitted by using an exponential decay function. As shown in Figure 2i, the GSB decay kinetics of PM6:BO-4Cl and PM6:HD-1:BO-4Cl films at 631 nm exhibited τ_1 values of (0.499 and 0.283 ps) and τ_2 values of (3.07 and 4.66 ps), respectively, where τ_1 and τ_2 correspond to the ultrafast exciton dissociation at the interface and the exciton diffusion in the domain, respectively. The shorter τ_1 indicates that the addition of HD-1 facilitates the exciton dissociation,^[58] which facilitates charge generation and a higher J_{SC} in the corresponding OSCs.

It is well established that a well-blended morphology of the active layer is essential for achieving high-efficiency OSCs. As we previously reported, higher-order (h00) diffraction peaks in the out-of-plane (OOP) direction are more prominent in the HD-1 **ADVANCED** SCIENCE NEWS _

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Figure 2. a) Light intensity dependence of J_{SC} and b) Light intensity dependence of V_{OC} of devices without and with HD-1. c) J_{ph} versus V_{eff} curves of the corresponding devices. d) TPC and e) TPV curves. f) The hole and electron mobilities of the BHJ obtained from single carrier devices. g,h) The TA spectra of blend films. i) Decay dynamics probed at 631 nm in blends under 800 nm excitation.

neat film compared to the PM6 and BO-4Cl neat film, suggesting the long-range order and high crystallinity with an orientation of the crystals parallel to the substrate surface.^[59] To investigate the underlying reasons for the improved performance of systems containing HD-1, grazing incidence wide-angle Xray scattering (GIWAXS) and atomic force microscopy-based infrared spectroscopy (AFM-IR) measurements were performed. Figure 3a-c display the GIWAXS 2D patterns of the blend films, along with line cut profiles along the out-of-plane (OOP) and inplane (IP) directions. The relevant parameters, including stacking distance and crystalline correlation lengths (CCLs), are summarized in Table S6 (Supporting Information). The HD-1-based ternary film exhibits a (010) peak at 1.69 Å⁻¹ (d = 3.72 Å) in the OOP direction, with a crystal coherence length (CCL) of 22.62 Å. In contrast, the binary film shows a (010) peak at 1.71 Å⁻¹ (d = 3.67 Å) in the OOP direction, with a CCL of 20.94 Å. Compared to the binary film, the ternary film, due to the enhanced crystallinity of HD-1, demonstrates a higher CCL value, indicating greater crystallinity and more orderly orientation,^[60] which is advantageous for charge dissociation and transport. Then, we employed AFM-IR measurements to investigate the fibril structures of the blend films (Figure 3d,e).^[61] The donors and BO-4Cl could be easily distinguished based on the presence or absence of the infrared vibration signal of C≡N group at 2216 cm⁻¹, as observed in the AFM-IR spectra, with the donors represented by blue and BO-4Cl by red. Notably, the ternary blend film exhibited a significantly superior fiber network structure compared to the binary film, which is crucial for supporting efficient charge transport.^[62] Additionally, a statistical size evaluation of the nanofibers was performed, revealing that the HD-1-based blend film had a smaller nanofiber size of 17.88 nm, compared to 24.20 nm for the binary blend film (Figure 3f; Figure S8, Supporting Information), indicating more interfaces between the donors and the acceptors for effectively exciton dissociation in ternary devices.

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Figure 3. a) 2D GIWAXS patterns for PM6:BO-4Cl and b) PM6:HD-1:BO-4Cl blend films. c) Corresponding 1D line-cuts of the corresponding films. d,e) The AFM-IR images of blend films. f) The histogram of the fibril width fitted with Gaussian distributions. g) DSC thermograms of HD-1, PM6, BO-4Cl and blends. h,i) In situ 2D UV-vis absorption mapping of PM6:BO-4Cl and PM6:HD-1:BO-4Cl active layers during thermal annealing treatment.

To further investigate the factors responsible for the distinct morphologies of the blends, differential scanning calorimetry (DSC) and in situ absorption measurements were performed. As previously reported, it was found that PM6 did not exhibit obvious endothermic or exothermic peaks, whereas a crystallization peak at 272 °C was observed for HD-1. When blended with BO-4Cl (in a 10:1 w/w ratio, which closely matches the experimental active layer composition), the crystallization peak of HD-1 disappeared, suggesting that HD-1 is well miscible with BO-4Cl. However, even with the addition of 10% HD-1 in PM6, the crystallization peak of HD-1 remained detectable (Figure 3g), indicating that the crystallization of both PM6 and HD-1 persisted and that the miscibility between the two is limited,^[63] which further supports the χ values obtained from the contact angle measurements. Additionally, the drying and crystallization dynamics of the active layers during thermal annealing treatment were analyzed through in situ absorption measurements to gain a deeper understanding of the working mechanisms (Figure 3h,i). When using CB as the processing solvent, BO-4Cl exhibited a strong tendency to self-aggregate, leading to phase separation and diffusion out of the donor phase. However, when HD-1, highly miscible with BO-4Cl, is added as the highly crystalline guest component into the active layer (PM6:BO-4Cl), it easily assembles and serves as nucleation sites to trigger heterogeneous nucleation,^[46] which effectively suppresses excessive aggregation of BO-4Cl during the film-forming process and achieve an active layer with ideal phase separation morphology.

To evaluate the adaptability of the ternary strategy for largearea devices, a 1 cm² device and a mini-module with an active area of 13.5 cm² were fabricated. As shown in Figure S12 (Supporting Information), the 1 cm² device demonstrated a high PCE of 18.09%, with a J_{SC} of 27.71 mA cm⁻² and an FF of 75.97%. The www.advancedsciencenews.com

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Figure 4. a) The schematic diagram labeling P1, P2, P3 of the OSC module (active area: 13.5 cm², six sub-cells in series). b) The *J*–V curve of the OSC module under AM 1.5 G. c) The *J*–V curve of the OSC module at 1000 lux indoor lighting. d) The equivalent schematic of the OSC-ESL system. e) The current variation in the electrical circuit and the calculated electricity consumption during the refreshing process. f) The photograph of the OSC-ESL system.

mini-module, which has an active area of 13.5 cm^2 and consists of six sub-cells connected in series, was fabricated using blade coating in air. A schematic diagram of the mini-module is shown in **Figure 4a**. The ternary mini-module exhibited an impressive PCE of 16.48%, with the V_{OC} of 5.04 V, J_{SC} of 4.44 mA cm⁻², and an FF of 73.57% (Figure 4b), demonstrating the potential of the ternary system for large-area printing processes.

Compared to 1-sun condition considered for outdoor OSCs, the intensity of indoor lights is much lower, commonly less than 1 mW cm⁻².^[64,65] Therefore, thick-film devices are ideal for maximizing the use of photocurrent and improving the efficiency of indoor organic photovoltaics.^[66,67] In this study, the addition of HD-1 effectively suppresses charge recombination in thickfilm devices, which contributes to better indoor photovoltaic performance. Under indoor light (1000 lux), the ternary module (13.5 cm²) achieves a maximum power output (P_{max}) of 45.48 μ W cm⁻² (Figure 4c). To further explore the practical application potential of the ternary system, we investigated the feasibility of integrating OSC modules with commercial electronic shelf labels (ESL).^[13] Two modules were connected in series as the energy source for rechargeable lithium batteries in ESL. The equivalent simple schematic of the OSC-ESL system is shown in Figure 4d. When the screen of ESL refreshed once, the current variation in the electrical circuit and the calculated electricity consumption during the refreshing process were shown in Figure 4e,f. The energy provided by OSC modules working at 1000 lux indoor lighting for an hour could refresh the ESL 46 times, as the calculated electricity consumption for refreshing ESL once is \approx 95 mJ, which shows excellent application prospects.

3. Conclusion

In summary, we introduced a small molecule donor HD-1 into the PM6:BO-4Cl system to modulate the active layer morphology and achieve high efficiencies in both devices and modules fabricated using the high-boiling solvent CB. Due to the good miscibility between BO-4Cl and HD-1, excessive aggregation of BO-4Cl was suppressed, resulting in optimized phase separation of the active layer when processed with CB. The improved morphology reduced charge recombination and enhanced the balance of carrier mobilities. As a result, the ternary device achieved an impressive PCE of 19.42%, with an FF of 80.00%. Furthermore, the device demonstrated good tolerance to variations in active layer thickness, with a 300-nm-thick film achieving a PCE exceeding 17%. Notably, a large-area module prepared by blade coating in air using CB achieved a PCE of 16.48%. Our study demonstrates that large-area modules incorporating a highly crystalline small molecule donor as the third component show significant potential for large-area organic photovoltaic applications.

Supporting Information

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

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

blade-coated module, high-boiling solvent, morphology modulation, organic solar cells

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