Enhancing Efficiency and Stability of Organic Solar Cells by UV Absorbent

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A new type of high boiling-point additive, UV absorbent benzophenone (BP), is reported which can simultaneously improve the efficiency and stability of fullerene and nonfullerene organic solar cells (OSCs). After the addition of BP, the power conversion efficiencies (PCEs) of nonfullerene OSCs based on FTAZ: ITIC-Th is increased from 8.5 to 9.4%, and is further increased to 10.3% by employing inverted geometry. Meanwhile, the photo-stability of nonfullerene OSC is improved. After illumination-aging, the OSCs with BP preserve 79% of the original PCEs, while the OSCs without additives or with 1,8-diiodooctane only preserve 65 and 58% of their original PCEs, respectively. In addition, BP can also work in fullerene-based OSCs. After the addition of BP, the efficiency and photo-stability of the OSCs based on PTB7-Th: PC₇₁BM are simultaneously enhanced.

As a promising technology for clean and renewable energy conversion, organic solar cells (OSCs) have attracted considerable attention in recent years because they present some advantages, such as low cost, light weight, flexibility, semitransparency and large-area fabrication.^[1–7] Much effort has been dedicated to enhancing the power conversion efficiencies (PCEs) of OSCs, such as new donor or acceptor materials synthesis, morphology control, and device engineering. Especially, morphology optimization of active layer is an effective and indispensable step for OSCs fabrication.^[8,9] In recent years, researchers have developed a number of methods to control the morphology of the polymer/

fullerene active layers, among which, the use of high boiling-point additives, such as 1,8-diiodooctane (DIO),^[10] chloronaphthalene (CN)^[11] and diphenyl ether (DPE),^[12] is the most widely used approach, which can control the solvent evaporation dynamics and thus significantly improve the PCEs.^[13–20] However, the residue of these high boiling-point solvent additives in OSCs is detrimental to the device stability,^[21–26] which limits the future industrial production of OSCs.^[27–29]

In recent 3 years, nonfullerene acceptor based OSCs have attracted increasing attention.^[30–44] Compared with fullerene acceptors, nonfullerene acceptors show some advantages, such as broad and strong

absorption and adjustable highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) energy levels. The nonfullerene OSCs show higher PCEs than their fullerene based counterparts. Similar to the fullerene based OSCs, high boiling-point additives (DIO, CN, DPE, etc.) also play an important role in fabrication of high efficiency nonfullerene OSCs.^[45–51] Thus, finding new additives, which can simultaneously enhance the efficiency and stability of nonfullerene OSCs, is timely and highly desired.

In this work, we used a new type of high boiling-point additive, benzophenone (BP), in fullerene and nonfullerene OSCs to control the morphology, leading to improved device efficiency and stability. Due to the high boiling point (305 °C), BP can control the solvent evaporation dynamics during the film formation, resulting in optimized morphology for higher PCEs. On the other hand, BP is a kind of low-cost and widely used ultraviolet absorbent (UV absorbent) in industrial manufacture, which can strongly and selectively absorb UV light (Figure S1, Supporting Information) and dissipate the high energy of UV light through innocuous low energy radiation. Since UV light contributes little to the photocurrent but damages OSCs due to the high energy, the residue of UV absorbent BP in the blended film is potentially beneficial to the photo-stability of OSCs. After optimization of the BP content, the PCE of nonfullerene OSC is increased from 8.5 to 9.4% (is further increased to 10.3% by employing inverted geometry), which is higher than that with DIO (9.0%). The photo-stability of nonfullerene OSC is improved: after 150 min illumination, the PCEs of the nonfullerene OSCs with BP preserve 79% of their original values, while those without additive or with DIO preserve 65 and 58% of their original values, respectively. In addition, BP can also work in fullerene-based

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OSCs. After the addition of BP, the efficiency and photo-stability of fullerene-based OSCs are simultaneously enhanced.

The structure of nonfullerene OSCs is indium tin oxide (ITO)/poly(3,4-ethylenedioxythiophene): poly(styrene sulfonate) (PEDOT: PSS)/FTAZ: ITIC-Th/calcium (Ca)/aluminum (Al), and the molecular structures of the wide-bandgap donor FTAZ^[52] and the narrow-bandgap acceptor ITIC-Th^[41] are shown in **Figure 1**. Because DIO is the most widely used and successful high boiling-point (332 °C) additive, we compared BP with DIO in terms of both efficiency and stability. **Figure 2** shows the *J*-*V* curves and external quantum efficiency (EQE) spectra of devices without or with additives under the illumination of an AM 1.5G solar simulator, 100 mW cm⁻², as well as the curves of PCEs versus illumination-aging time under nitrogen atmosphere. The photo-stability tests were carried out under continuous 150 min illumination by an AM 1.5G solar simulator, with cooling fans to eliminate the interference of heat. The optimized content of BP is 7% of the total weight of donor FTAZ and acceptor ITIC-Th, while the optimized content of DIO is 0.25% of the processing solvent volume. The average and best device characteristics are summarized in **Table 1** (the average data are calculated from 10 individual devices, and the PCE/PCE₀ is defined as the ratio of PCEs after and before the photo-stability test). By employing BP into the devices, the short circuit current density (*J*_{SC}) and fill factor (FF) are increased, while the open circuit voltage (*V*_{OC}) is not affected. The average PCE is increased from 8.2 to 9.1%. In comparison, devices with DIO show average PCE of 8.7%, lower than that of devices with BP. In addition, BP was also employed in fullerene-based OSCs (PTB7-Th: PC₇₁BM) to investigate its generality. With the addition of 7% BP into the devices, the average PCE is increased from 7.4 to 8.9%. In comparison, devices with 3% DIO show average PCE of 8.3%, lower than that of devices with BP.

As shown in Figure 2b, the trend of EQE is similar to *J*_{SC}. According to the absorption spectra of donor and acceptor materials, the EQE under 600 nm is mainly attributed to FTAZ, while the EQE in 600–800 nm is attributed to ITIC-Th. After the addition of 7% BP, the EQE related to both donor and acceptor parts are obviously enhanced. To evaluate the accuracy of the

Figure 1. Molecular structures of FTAZ, PTB7-Th, ITIC-Th, PC₇₁BM, and BP.

Figure 2. Photovoltaic characteristics and photo-stability tests of OSCs based on ITO/PEDOT: PSS/FTAZ: ITIC-Th/Ca/Al without additives (w/o), with DIO (w/ DIO), and with BP (w/ BP). a) *J*-*V* curves and b) EQE spectra of devices. c) Photo-stability curves of devices under continuous illumination-aging for 150 min (AM 1.5G solar simulator, 100 mW cm⁻²).

photovoltaic results, the *J*_{SC} values are calculated from integration of the EQE spectra with the AM 1.5G reference spectrum. The calculated *J*_{SC} is similar to *J*-*V* measurement (the average error is 3.1%, Table 1).

Since both BP and DIO possess high boiling points, some of them would remain in the active layers. The residue of DIO was verified before by fourier transform infrared (FTIR).^[21] Similarly, by comparison of FTIR spectra of films without and with BP

Table 1. Average and best device data based on FTAZ: ITIC-Th or PTB7-Th: PC₇₁BM without or with additives and their photo-stability after 150 min illumination-aging

Acceptor	Additive	V _{OC} (V)	J _{SC} (mA cm ⁻²)	Calc. J _{SC} (mA cm ⁻²)	FF (%)	PCE (%)		PCE/PCE ₀ (%)
						Average	Best	
ITIC-Th	–	0.91 ± 0.01	14.0 ± 0.2	13.6	64.5 ± 0.8	8.2 ± 0.2	8.5	65
ITIC-Th	DIO	0.91 ± 0.01	14.4 ± 0.3	13.9	65.8 ± 0.9	8.7 ± 0.2	9.0	58
ITIC-Th	BP	0.91 ± 0.01	15.2 ± 0.2	14.6	66.0 ± 1.0	9.1 ± 0.2	9.4	79
PC ₇₁ BM	–	0.81 ± 0.01	14.0 ± 0.2	13.6	65.3 ± 0.8	7.4 ± 0.2	7.6	73
PC ₇₁ BM	DIO	0.81 ± 0.01	15.3 ± 0.3	14.8	67.2 ± 0.9	8.3 ± 0.4	8.9	63
PC ₇₁ BM	BP	0.81 ± 0.01	16.2 ± 0.2	15.9	68.0 ± 0.6	8.9 ± 0.3	9.3	85

(Figure 3), it is obvious that BP remains in the film, which is also supported by UV absorption test (Figure S1, Supporting Information) and gas chromatography-mass spectrometer (GC-MS) test (Figures S2 and S3, Table S1, Supporting Information). After illumination-aging for 150 min, FTAZ: ITIC-Th devices without any additives, with DIO and with BP exhibit average PCEs of 5.3, 5.1, and 7.2%, respectively, preserving 65, 58, and 79% of original PCE values (Figure 2c). After illumination-aging at elevated temperature of 60 °C for 150 min, the devices without additive, with DIO, and with BP exhibit PCEs of 4.8%, 4.3%, 7.0%, preserving 59%, 49%, 77% of their original PCE values, respectively (Figure S4, Supporting Information). Similar to nonfullerene devices, after illumination-aging for 150 min, PTB7-Th: PC₇₁BM devices without any additives, with DIO and with BP preserve 73, 63, and 85% of original PCE values, respectively. The addition of BP can significantly enhance the photo-stability of OSCs.

High boiling-point additives can control the morphology of active layer during film formation. Figure 4 displays transmission electron microscopy (TEM) images of FTAZ: ITIC-Th blended films without or with additives to investigate the effects of additives on morphology before and after illumination-aging. After illumination-aging for 150 min, the morphology of films without additives and with DIO are changed, while the film with BP remains unchanged. It has been reported that the residual DIO in the blended film will be decomposed under illumination,

and the decrease in DIO amount will change the morphology of active layer.^[21,25,53] Different from DIO, as a common industrial UV absorbent, the residual BP in blended film is stable under illumination, and thus the active layer can maintain the original morphology. On the other hand, the donor and acceptor materials would suffer from photochemical reaction under illumination of the high-energy UV light, leading to changes of their molecular structures.^[21] As a result, the miscibility of donor and acceptor materials would be changed, which accordingly affects the phase separation of active layer. The addition of BP into the blended film may prevent the molecular structures of donor and acceptor materials from destroying by UV light, resulting in stabilized morphology of the active layer.

Figure 5a–f shows the 2D grazing-incidence small-angle X-ray scattering (GISAXS) patterns of FTAZ: ITIC-Th blended films without or with additives before and after illumination-aging. The in-plane scattering intensity profiles are shown in Figure 5g. By fitting them with the models adopted in previous studies,^[54,55] we could roughly estimate the average domain sizes of intermixing amorphous phases, FTAZ domains and ITIC-Th domains, which are summarized in Table S2 (Supporting Information). The FTAZ domains remain almost unchanged (≈ 4 nm) for all the films, as manifested by the shoulder appeared at $q_r \approx 0.08 \text{ \AA}^{-1}$. However, the acceptor and intermixing domain sizes suffer from large changes after illumination-aging for the films without additive or with DIO, consistent with the observed significant performance deterioration of devices made with these two types of films. Impressively, the films with BP exhibit unprecedented morphology stability under illumination with almost unchanged donor, acceptor and intermixing domain sizes, in support of the observed stable device performance of FTAZ: ITIC-Th blended films with BP under illumination-aging.

Since BP is capable of controlling and stabilizing the morphology of active layer, which may affect charge transport, the hole mobility and electron mobility of FTAZ: ITIC-Th blended films without or with additives before and after 150 min illumination-aging were measured by space charge limited current (SCLC) method.^[56] Hole-only and electron-only diodes were fabricated using the architectures: ITO/PEDOT: PSS/active layer/Au for holes and Al/active layer/Al for electrons. As shown in Figure S5 and Table S3 (Supporting Information), addition of DIO or BP can increase the hole mobility from 2.1×10^{-5} to $7.5 \times 10^{-5} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, while slightly affect the electron mobility. After illumination-aging for 150 min, the blended films exhibit little changes of electron mobilities (from 1.5×10^{-5} to

Figure 3. FTIR spectra of FTAZ films without additives and with BP. The characteristic peak of —C=O— at 1652 cm^{-1} demonstrates BP would remain in the film. These films have the same thickness. ITIC-Th is not used to eliminate its interference of —C=O— functional group.

Figure 4. TEM images of FTAZ:ITIC-Th blended films without or with additives before (a–c) and after (d–f) 150 min illumination-aging. a and d) Without additives. b and e) With DIO. c and f) With BP. The scale bar is 100 nm.

Figure 5. 2D GISAXS images of FTAZ:ITIC-Th blended films without or with additives before (a–c) and after (d–f) 150 min illumination-aging. a and d) Without additives. b and e) With DIO. c and f) With BP. g) GISAXS in-plane scattering intensity profiles with fitting results.

$1.0 \times 10^{-5} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for film without additives, unchange of $1.2 \times 10^{-5} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ for films with DIO or BP). However, the hole mobilities of films without additives and with DIO considerably decrease from 2.1×10^{-5} to $9.2 \times 10^{-6} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, and from 7.5×10^{-5} to $9.2 \times 10^{-6} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, respectively. In contrast, attributed to the stabilized morphology, the blended film with BP experiences smaller change in hole mobility (from 7.5×10^{-5} to $5.2 \times 10^{-5} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$).

To investigate the influences of additives on the vertical phase separation of FTAZ: ITIC-Th blended films, X-ray photoelectron spectroscopy (XPS) was used to measure the ratio of atoms at the top surface of active layer (Figure S6, Supporting Information). Since among the donor FTAZ, acceptor ITIC-Th and additives DIO or BP, nitrogen (N) and sulfur (S) are presented only in FTAZ and ITIC-Th, the spectral lines of N 1s ($\approx 400 \text{ eV}$) and S 2p ($\approx 160 \text{ eV}$) are attributed to both FTAZ and ITIC-Th. One FTAZ repeated unit contains three N atoms and four S atoms, while one ITIC-Th molecule contains four N atoms and eight S atoms. From the N/S ratio, the polymer weight content at the top surface can be calculated: 66, 88, and 79% for blended films without additives, with DIO and with BP, respectively (details in the Supporting Information). In the blended films, the donor FTAZ tends to aggregate on the top surface, due to its relatively low surface energy, which is proven by the results of contact angle (CA) measurement ($101.6 \pm 1.0^\circ$ for FTAZ, $44.7 \pm 0.6^\circ$ for ITIC-Th, Figure S7, Supporting Information). High boiling-point additive BP can control the solvent evaporation dynamics, similar to DIO, leading to more aggregation of FTAZ on the top surface. As higher content of polymer donor on the top surface is beneficial to hole extraction by the top electrode, we employed inverted geometry (ITO/zinc oxide (ZnO)/FTAZ: ITIC-Th/molybdenum trioxide (MoO_3)/silver (Ag)) in the nonfullerene OSCs, to further explore the potential of BP for increasing PCEs. With addition of 7% BP, the device exhibits PCE of 10.3% (Table S4 and Figure S8, Supporting Information). The J_{SC} is 17.8 mA cm^{-2} , which is higher than that of the conventional geometry (15.2 mA cm^{-2}).

In summary, a new type of additive, UV absorbent BP, is employed in nonfullerene OSCs based on FTAZ: ITIC-Th to simultaneously enhance the PCE and stability. BP possesses two crucial characteristics: high boiling point and absorption of UV light. Due to the high boiling point, BP is capable of controlling the solvent evaporation dynamics during film formation, leading to optimized morphology of active layer. Due to increase in J_{SC} and FF, the PCE of FTAZ: ITIC-Th based devices with BP is enhanced by a factor of 11% relative to that without additives. An even higher PCE of 10.3% is achieved by using inverted geometry. On the other hand, thanks to the strong absorption in UV spectral region, residual BP in active layer can effectively alleviate photo-damages and stabilize the morphology, resulting in improved photo-stability of OSCs. After 150 min illumination-aging, devices with BP preserve 79% of the original PCE values, while those without additives and with DIO can only preserve 65 and 58% of their original PCEs. In addition, BP can also work in fullerene-based OSCs. After the addition of BP in PTB7-Th/ PC_{71}BM based OSCs, the efficiency and photo-stability are simultaneously enhanced. Our results demonstrate that additive BP presents an effective and economic approach to fullerene and nonfullerene OSCs with high efficiency and good stability.

Experimental Section

Unless stated otherwise, solvents and chemicals were obtained commercially and used without further purification. FTAZ^[52] and ITIC-Th^[40] were synthesized according to our previously reported procedures. PTB7-Th was purchased from 1-Materials Inc. PC_{71}BM was purchased from Solarmer Inc. BP, DIO, and o-dichlorobenzene (DCB) were obtained from J&K Chemical Inc.

Organic solar cells were fabricated with the structure: ITO/PEDOT: PSS/active layer/Ca/Al for regular geometry, and ITO/ZnO/active layer/ MoO_3 /Ag for inverted geometry. The ITO glass (sheet resistance = $10 \Omega \square^{-1}$) was pre-cleaned in an ultrasonic bath of acetone and isopropanol, and treated in ultraviolet-ozone chamber (Jelight Company, USA) for 20 min. For regular geometry, a thin layer (35 nm) of PEDOT: PSS (Baytron PVP AI 4083, Germany) was spin-coated onto the ITO glass and baked at 150°C for 20 min. A mixture of FTAZ and ITIC-Th was dissolved in DCB solvent (1: 1, 18 mg mL^{-1} in total) with stirring overnight. A mixture of PTB7-Th and PC_{71}BM was dissolved in DCB solvent (1: 1.5, 25 mg mL^{-1} in total) with stirring overnight. BP was also dissolved in DCB solvent (100 mg mL^{-1}). To fabricate OSCs with additives, appropriate volume of BP solution or DIO was added to the solution of active materials. Afterwards, the solutions were spin-coated on the PEDOT: PSS layer to form a photoactive layer. The thicknesses of the active layers were ranging from 80 to 100 nm, as measured by DektakXT (Bruker). A Ca (ca. 20 nm) and Al layer (ca. 80 nm) was then evaporated onto the surface of the photoactive layer under vacuum (ca. 10^{-5} Pa) to form the negative electrode. For inverted geometry, a thin layer (30 nm) of ZnO precursor solution was spin-coated onto the ITO glass and baked at 200°C for 60 min. The fabrication method of active layer in inverted geometry is the same as that in regular geometry. A MoO_3 (ca. 10 nm) and Ag layer (ca. 100 nm) were then evaporated onto the surface of the photoactive layer under vacuum (ca. 10^{-5} Pa) to form the back electrode. The active area of the device was 4 mm^2 . The J - V curve was measured using a computer-controlled B2912A Precision Source/Measure Unit (Agilent Technologies). An XES-70S1 (SAN-EI Electric Co., Ltd.) solar simulator (AAA grade, $70 \times 70 \text{ mm}^2$ photo beam size) coupled with AM 1.5G solar spectrum filters was used as the light source, and the optical power at the sample was 100 mW cm^{-2} . A $2 \times 2 \text{ cm}^2$ monocrystalline silicon reference cell (SRC-1000-TC-QZ) was purchased from VLSI Standards Inc. The EQE spectrum was measured using a Solar Cell Spectral Response Measurement System QE-R3011 (Enlitech Co., Ltd.). The light intensity at each wavelength was calibrated using a standard single crystal Si photovoltaic cell. Photo-stability tests of solar cells were carried out under AM 1.5 G illumination for 150 min, in an open-circuit status without a shadow mask, with cooling fans to avoid the interference of heat or at elevated temperature of 60°C . These tests were carried out in glove box.

UV-vis absorption spectrum was recorded on a JASCO V-570 spectrophotometer. The testing samples (solution) for the absorption spectra were prepared as below: 1) FTAZ: ITIC-Th blend films without or with BP were fabricated under the same condition as the devices described above; 2) These samples were treated under the vacuum condition, which was same as that of the devices; 3) The blend films without or with BP and pure BP powder

were respectively dissolved and diluted in chloroform to achieve appropriate concentrations for the absorption tests. To prepare samples for FTIR tests, the polymer solutions without or with BP were firstly spin coated on potassium bromide (KBr) with the same thickness. Then these samples were treated under the vacuum condition before FTIR tests. The vacuum treatment process of FTIR samples was same as that of the devices. FTIR studies were performed using a Bruker Tensor 27 FTIR spectrometer. The sample for GC-MS test was prepared as below: 1) a DCB solution of FTAZ:ITIC-Th:BP (1:1:0.14, w/w/w) was spin-coated on PEDOT:PSS layer to form the active layer; 2) This sample was treated under the vacuum condition, which was same as that of the devices; 3) The active layer was soaked with 50 μ L methanol on the surface for 60 s. The methanol solution was analyzed with GC-MS. The transmission electron microscopy (TEM) characterization was carried out on a JEM-2100 transmission electron microscope operated at 200 kV. The samples for the TEM measurements were prepared as follows: the active layer films were spin-casted on ITO/PEDOT:PSS substrates, and then substrates with the active layers were submerged in deionized water to make the active layers float onto the air-water interface. Then, the floated films were picked up on unsupported 200 mesh copper grids for the TEM measurements. The grazing incidence small-angle X-ray scattering measurements (GISAXS) were carried out at BL23A1 of National Synchrotron Radiation Research Center, Hsinchu. The energy of the X-ray source was set to 10 keV (wavelength of 1.24 Å) and the incident angle was 0.15°. Hole-only or electron-only diodes were fabricated using the architectures: ITO/PEDOT:PSS/active layer/Au for holes and Al/active layer/Al for electrons. Mobilities were extracted by fitting the current density–voltage curves using the Mott–Gurney relationship (space charge limited current). XPS was performed on the Thermo Scientific ESCALab 250Xi using 200 W monochromated Al K α radiation. The 500 μ m X-ray spot was used for XPS analysis. The base pressure in the analysis chamber was about 3×10^{-10} mbar. Typically, the hydrocarbon C1s line at 284.8 eV from adventitious carbon was used for energy reference. Static contact angles were measured on a dataphysics OCA20 contact-angle system at ambient temperature (the test liquid is water).

Supporting Information

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

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