PHOTONICS Research

19.34 cm² large-area quaternary organic photovoltaic module with 12.36% certified efficiency

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Received 2 December 2020; revised 22 December 2020; accepted 23 December 2020; posted 24 December 2020 (Doc. ID 416229); published 19 February 2021

In this study, a quaternary blending strategy was applied in the fabrication of organic photovoltaic devices and large-area modules. As a result, the ultimate quaternary organic solar cells (OSCs) deliver 16.71% efficiency for small-area devices and 13.25% for large-area (19.34 cm²) modules (certified as 12.36%), which is one of the highest efficiencies for organic solar modules to date. Our results have proved the synergistic effects of multiple components in OSCs, providing an effective strategy for achieving high-performance organic photovoltaic devices and modules. © 2021 Chinese Laser Press

https://doi.org/10.1364/PRJ.416229

1. INTRODUCTION

Donor-acceptor organic solar cells (OSCs) have achieved tremendous progress since the reports of bulk-heterojunction (BHJ) organic solar cells [1]. Early in its development, fullerenes and their derivatives led to an era of OSCs due to their excellent electron mobility, large electron acceptability, and isotropy of charge transport [2–4]. However, the inherent huge energy losses and weak photon-harvesting capacities of fullerenes limited their further applications [5,6]. Compared with fullerene acceptors, the non-fullerene acceptors (NFAs) usually have strong absorption in the near-infrared region (NIR), and their energy levels can be easily adjusted to match the donors, which have been proved to promote the power conversion efficiencies (PCEs) of OSCs [7–12]. Driven by the vigorous development of materials science, the PCEs of single-junction OSCs have recently exceeded 17% [13-21]. Despite the impressive achievements made by NFAs, the single NFA usually suffers from incomplete absorption of the solar spectrum and relatively low charge mobility compared with perovskite components [22]. In order to make full use of the advantages of fullerene and NFAs, multicomponent blending is recommended to construct multiple devices. The incorporation of fullerene and NFAs into the BHJ layer provides a promising approach to improve the photovoltaic performance of OSCs.

The recent rapid progress of OSC materials, especially NFAs with variable molecular structures and tunable energy levels,

2327-9125/21/030324-07 Journal © 2021 Chinese Laser Press

enables a large material pool for realizing the potential of multicomponent BHJ solar cells [23-29]. Li et al. [30] earlier reported an NFA-based binary system BDB-T:ITIC mixed with binary fullerenes as acceptor additives. Consequently, the optical band gaps of the NFA-blended films were reduced and the carrier transport processes were enhanced. As a result, the open-circuit voltage $(V_{\rm OC})$, the short-circuit currentdensity (I_{SC}) , and the fill factor (FF) were simultaneously improved, promoting an ultimate PCE up to 12.8%. Bi et al.[31] developed an individual nanostructure optimized quaternary system based on two donors and two acceptors. High crystallinity DR3TBDTT was dispersed in PTB7-Th to enhance the domain purity, while PC71BM was used as phase modifier to promote favorable FOIC packing. Thus, the nanoscale morphologies of the donors and the acceptors were optimized individually, which helped to increase carrier mobilities and suppress monomolecular recombination, thereby contributing to a champion PCE of 13.51%. Recent work from Ma et al. [32] proposed an efficient quaternary system combining a basic binary blend including fullerene and NFA. The third component, Br-ITIC, was added to maximize the photon harvesting, and PC71BM was used as the fourth component to optimize the molecular arrangement and phase separation of the active layer, leading to a PCE of 16.8%. Therefore, multicomponent blending is an effective strategy to promote the PCEs of OSCs. However, quaternary or ternary

blending has rarely been applied in large-area photovoltaic modules.

In this work, a quaternary blending strategy was used in the fabrication of small-area photovoltaic devices and large-area photovoltaic modules. We have explored a quaternary system, including one polymer donor PM6, one fullerene acceptor PC71BM, and two NFAs Y6 and ITIC. The PC71BM was added as the third component in the PM6:Y6 system to optimize the charge transport. The ITIC was further added as the fourth component to maximize the photon harvesting. As a result, a champion PCE of 16.71% was achieved in the quaternary device after an optimization of photovoltaic parameters. Furthermore, a certified PCE of 12.36% was achieved for the large-area (19.34 cm^2) module, which is one of the highest efficiencies for inverted organic solar modules to date. The excellent photovoltaic performance demonstrates the great potential of using multicomponent organic solar modules for practical applications.

2. EXPERIMENT

A. Materials

The PM6, Y6, ITIC, and $PC_{71}BM$ were purchased from Solarmer Materials Inc. The ZnO precursor solution was prepared by dissolving zinc acetate dehydrate and ethanolamine in the solution of 2-methoxyethanol with a concentration of 0.5 mol/L.

B. Device Fabrication

The multiple OSCs were constructed with indium tin oxide (ITO)/ZnO/active layer/MoO_x/Ag. For small-area device fabrication, the ZnO precursor solution was spin-coated on the oxygen plasma-pretreated ITO at 3000 r/min and baked at 200°C for 1 h in air. PM6, Y6, ITIC, and PC₇₁BM were dissolved in chloroform with different mass ratios. The mass concentration of the polymer was kept at 7 g/L. Chloronaphthalene (0.8%) was added to optimize the phase separation. The blend solutions were then spin-coated onto the ZnO layer at an optimized speed of 2400 r/min, obtaining the thickness of ~100 nm, and were subsequently annealed at 50°C for 10 min. Finally, a 10 nm thick MoO_x layer was deposited as the anode interlayer by thermal evaporation under less than 5×10^{-5} Pa, followed by deposition of 80 nm of Ag as the top electrode.

For large-area module fabrication, the modules were fabricated with a similar architecture to the small-area OSCs. The patterned ITO-coated glass substrates (6 cm × 6 cm with P1 scribe) were spin-coated with a ZnO precursor solution at 3000 r/min and baked at 200°C for 1 h in air. The quaternary blend solution (PM6:Y6:ITIC:PC₇₁BM = 1:1.15:0.05:0.2) was prepared with a polymer weight concentration of 9 g/L. Chloronaphthalene (0.8%) was added as additive. The quaternary solution was spin-coated on the ZnO layer at 5000 r/min in N₂ atmosphere, and the films were subsequently thermally annealed at 50°C for 10 min. Then a 10 nm thick MoO_x layer was deposited as the anode interlayer by thermal evaporation under less than 5×10^{-5} Pa. The series connection is realized with the typical P1-P3 laser

(532 nm, 10 ns) patterning method, as shown later in this paper, and the geometrical filling factor is 95.5%.

C. Characterization

The density-voltage (*J-V*) curves were measured by a Keithley 2400 source meter under AM 1.5 G light (100 mW·cm⁻²) from an SAN-EI electric solar simulator. The light intensity was calibrated using a standard silicon solar cell. The typical small-device area of 0.063 cm² is defined by a shadow mask. For the large-area module, the total illumination area is 19.34 cm², and the external quantum efficiency (EQE) was recorded by QE-R from Enli Technology. The UV-vis absorption spectra were measured by a Agilent Carry 5000 UV-vis spectrometer. Atomic force microscopy (AFM) images of the sample surfaces were obtained on a Cypher S atomic force microscope.

3. RESULTS AND DISCUSSION

Figure 1(a) depicts the chemical structures of the four photovoltaic materials. The UV-vis absorption spectra of neat PM6, Y6, ITIC, and PC₇₁BM films are displayed in Fig. 1(b), covering the absorption range of 500–900 nm in the solar spectrum. The OSCs with an inverted structure of ITO/ZnO/active layer/MoO_x/Ag were fabricated to evaluate the photovoltaic performance of the multiple devices, where the active layer consists of PM6, Y6, ITIC, and PC71BM with different mass ratios. The current density-voltage (J-V) curves of the multiple OSCs under illumination of AM 1.5 G (100 mW ⋅ cm⁻²) are shown in Fig. 1(c), and the detailed photovoltaic parameters are summarized in Table 1. A champion PCE of 16.71% was achieved in the quaternary device (PM6:Y6:ITIC: $PC_{71}BM = 1:1.15:0.05:0.2$), which was significantly higher than those in the binary (15.47%) and the ternary ones (16.29%). Based on the binary PM6:Y6 system, PC71BM was added as the third component, leading to large increase of J_{SC} and FF. The significant improvement in photovoltaic performance benefits from the more balanced charge transport, which is caused by the excellent electron mobility of the fullerene [33,34]. Then ITIC was added as the fourth component to optimize the photovoltaic parameters of the quaternary devices. Notably, the $V_{\rm OC}$ of the quaternary devices can be adjusted near linearly by the increase of ITIC content. With the appropriate addition of the ITIC component, the PCE is further enhanced to 16.71% with J_{SC} and FF slightly increased to 25.63 mA· cm⁻² and 76.22%, respectively. The EQE curves of the multiple OSCs are illustrated in Fig. 1(d). The optimal quaternary device exhibits a higher integrated J_{SC} of 24.50 mA· cm⁻² than the reference values of the corresponding binary and ternary devices, which matches well with the J_{SC} values calculated from the J-V curves.

To investigate the surface morphology of the active layer films, we performed the morphology characterization using the AFM. As shown in Fig. 2, all the blend films exhibit uniform morphology with relatively low root-mean-square surface roughness (R_q) values, showing the good miscibility among the four components. The binary PM6:Y6 film possesses an R_q of approximately 1.18 nm, and it is decreased to 0.83 nm after the addition of the third component PC₇₁BM. When adding an



Fig. 1. (a) Chemical structures and (b) normalized absorption spectra of PM6, Y6, ITIC, and PC₇₁BM. (c) *J-V* curves and (d) EQE spectra of the multiple OSCs.

Table 1. Photovoltaic Parameters of the Multiple OSCs

PM6:Y6:ITIC:PC71BM	$V_{\rm OC}$ (V)	$J_{\rm SC}({\rm mA}{\cdot}{\rm cm}^{-2})$	$J_{\rm SC, EQE}^{a}$ (mA·cm ⁻²)	FF (%)	PCE (%)
1:1.4:0:0 ^b	0.844 ± 0.005	24.48 ± 0.27	23.86	74.88 ± 0.63	15.47 ± 0.23
1:1.2:0:0.2	0.844 ± 0.005	25.44 ± 0.34	24.26	75.85 ± 0.31	16.29 ± 0.21
1:1.15:0.05:0.2	0.856 ± 0.011	25.63 ± 0.42	24.50	76.22 ± 1.18	16.71 ± 0.33
1:1.1:0.1:0.2	0.856 ± 0.011	25.46 ± 0.08	24.34	75.06 ± 1.25	16.35 ± 0.22
1:1:0.2:0.2	0.867 ± 0.011	24.57 ± 0.45	23.92	73.13 ± 0.61	15.57 ± 0.29
1:0.8:0.4:0.2	0.878 ± 0.011	23.95 ± 0.43	22.75	69.00 ± 0.48	14.51 ± 0.37
1:0.6:0.6:0.2	0.911 ± 0.005	22.33 ± 0.37	21.26	61.34 ± 0.25	12.48 ± 0.36
1:0.4:0.8:0.2	0.944 ± 0.010	20.04 ± 0.26	19.10	58.48 ± 1.32	11.07 ± 0.11
1:0.2:1:0.2	0.967 ± 0.011	17.04 ± 0.17	16.20	53.15 ± 0.89	8.76 ± 0.16
1:0:1.2:0.2	0.968 ± 0.011	13.52 ± 0.33	12.87	47.46 ± 0.48	6.20 ± 0.17

"The integrated J_{SC} calculated from the EQE curves of the multiple devices.

^bThe multiple devices based on PM6:Y6:ITIC:PC₇₁BM (area = 0.063 cm²). The average PCE values were obtained from over 20 devices.

appropriate amount of the fourth component ITIC (1:1.15:0.05:0.2), the R_q is further optimized as 0.79 nm. The well-formed domain sizes and phase separation are beneficial for charge separation, which is consistent with *J*-*V* measurement. As the ITIC content is further increased, the R_q starts to increase instead, indicating a negative effect on the morphology caused by excess ITIC content.

To evaluate the exciton dissociation and charge extraction processes in the multiple OSCs, the photocurrent density $(J_{\rm ph})$ versus the effective voltage $(V_{\rm eff})$ was carried out, where $J_{\rm ph}$ and $V_{\rm eff}$ were obtained by $J_{\rm light}$ - $J_{\rm dark}$ and V_0 - $V_{\rm bias}$, respectively. The $J_{\rm ph}$ - $V_{\rm eff}$ curves of the multiple devices are shown in Fig. 3. The $J_{\rm ph}$ increases rapidly and eventually reaches a saturated state, indicating the sufficient dissociation and collection of the photogenerated charges. The $J_{\rm ph}/J_{\rm sat}$ values under short-circuit current were calculated to evaluate the exciton dissociation of the multiple devices, where the $J_{\rm sat}$ corresponds with the saturated photocurrent density at $V_{\rm eff} = 2$ V. As summarized in Table 2, the optimal quaternary device (1:1.15:0.05:0.2) exhibits the highest value of 0.961, which is slightly higher than the corresponding binary (0.953) and ternary devices (0.959), indicating more efficient exciton dissociation and charge collection. The appropriate addition of PC₇₁BM and ITIC in the binary PM6:Y6 blend can provide more donor/acceptor interfaces for favorable exciton dissociation and charge transport, leading to increased $J_{\rm SC}$ in multiple devices.

Based on the high performance of the quaternary OSCs, we manufactured a large-area photovoltaic module [Fig. 4(a)] with the optimal condition (1:1.15:0.05:0.2).



Fig. 2. AFM height images and R_q values of the multiple OSCs. PM6:Y6:ITIC:PC₇₁BM equals (a) 1:1.4:0:0, (b) 1:1.2:0:0.2, (c) 1:1.15:0.05:0.2, (d) 1:1.1:0.1:0.2, (e) 1:0.6:0.6:0.2, and (f) 1:0:1.2:0.2.

The monolithic module is designed and realized by series connection of seven subcells. Laser engraving, known as the P1, P2, P3 laser patterning method, is used to connect the anode of one cell and the cathode of the subsequent cell. A realistic image of the module is shown in Fig. 4(b). The total illumination area is 19.34 cm², with geometrical filling factor of 95.5%. Figure 4(c) depicts the *J*-*V* curve of the large-area module. A PCE of 13.25% was realized, with a $V_{\rm OC}$ of 6.024 V, a $J_{\rm SC}$ of 3.11 mA·cm⁻², and an FF of 70.7%.



Fig. 3. $J_{\rm ph}$ - $V_{\rm eff}$ curves of the multiple OSCs.

It is noted here the efficiency is calculated based on the total illumination area, including both the photoactive area and the inactive connecting area, as defined by a photomask. A certified PCE of 12.36% was obtained, with a $V_{\rm OC}$ of 6.06 V, a $J_{\rm SC}$ of 3.07 mA·cm⁻², and an FF of 66.45% (Appendix A, Fig. 5). The PCE is the highest value in the inverted large-area module to date, and it is comparable to the performance of the state-of-the-art conventional modules (Table 3) [35–39]. The large-area photovoltaic module can even charge a cellphone under an indoor LED lamp [Fig. 4(d) and Visualization 1], revealing a promising commercialization.

Table 2.	Photovoltaic	Parameters	Calculated	from	the
$J_{\rm ph} - V_{\rm eff}$	Curves of the	Multiple OS	Cs		

PM6:Y6:ITIC:PC ₇₁ BM	$J_{\rm sat}$ (mA· cm ⁻²)	$J_{\rm ph} \ ({ m mA}\cdot{ m cm}^{-2})$	J _{ph} /J _{sat} (%)
1:1.4:0:0	25.68	24.48	95.3
1:1.2:0:0.2	26.53	25.44	95.9
1:1.15:0.05:0.2	26.67	25.63	96.1
1:1.1:0.1:0.2	26.62	25.46	95.6
1:1:0.2:0.2	25.83	24.57	95.1
1:0.8:0.4:0.2	25.27	23.95	94.8
1:0.6:0.6:0.2	23.93	22.33	93.3
1:0.4:0.8:0.2	21.82	20.04	91.8
1:0.2:1:0.2	18.74	17.04	90.9
1:0:1.2:0.2	15.66	13.52	86.3

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Fig. 4. (a) Device architecture, (b) realistic image, and (c) *J*-*V* curve of the large-area organic photovoltaic module. (d) The cellphone charged by the large-area organic photovoltaic module (see Visualization 1).



Fig. 5. Independent certification result of the large-area module based on the PM6:Y6:ITIC:PC₇₁BM (1:1.15:0.05:0.2) device from the Shanghai Institute of Microsystem and Information Technology, Chinese Academy of Sciences, confirming a high PCE of 12.36% (Certificate No. 19TR120402) (area = 19.34 cm^2).

Table 3.	Partial PCE C	omparison o	of the Large-	area Modules	with Areas	over 10 cm	² in Recent	Years

Modules	Configuration	Area (cm ²)	PCE (%)	Refs.
PTB7-Th:PCBM:COi8DFIC	Inverted	30	8.6	[35]
TPD-3 F:IT-4 F	Inverted	20.4	10.08 (certified)	[36]
PBDB-T:ITIC	Inverted	15	8.90	[37]
PM6:DTY6	Conventional	18	13.98 (certified)	[38]
PM6:Y6	Conventional	11.52	11.86	[39]
PM6:Y6:ITIC:PC71BM	Inverted	19.34	12.36 (certified)/13.25 (in house)	This work

4. CONCLUSION

In summary, we successfully fabricated a series of high-performance multiple OSCs. Fullerene and NFA (namely, $PC_{71}BM$ and ITIC) were sequentially added to the binary system to optimize the photovoltaic properties of the multiple blend films. With the strategy of integrating the advantages of multiple components, an optimized PCE up to 16.71% was obtained. Based on the exploration of quaternary devices, a large-area photovoltaic module was manufactured with a PCE of 13.25% (certified as 12.36%). The excellent performance of photovoltaic devices and large-area modules proves the synergy of multiple components, paving a way to the commercial application of the high-performance photovoltaic cells.

APPENDIX A

The main text presents the photovoltaic parameters of the large-area module measured in house. Here shows the independent certification result of the large-area module (Fig. 5). A certified PCE of 12.36% was obtained.

Funding. National Key Research and Development Program of China (2017YFA0207700); National Key Research and Development Program of Zhejiang Province (2018C04SA170313); Outstanding Youth Fund of Natural Science Foundation of Zhejiang (LR18F050001); National Natural Science Foundation of China (61705194, 61804134).

Disclosures. The authors declare no conflicts of interest.

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