Efficient Polymer Solar Cells with Open-Circuit Voltage of 1.01 V and Power Conversion Efficiency of 8.09%

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ABSTRACT: A series of polymer solar cells (PSCs) were prepared with different solvent additive 1-chloronaphthalene (CN) doping volume ratio to adjust the phase separation of active layers. The optimized PSCs exhibit a power conversion efficiency (PCE) of 8.09%, along with an open-circuit voltage of 1.01 V, a short circuit current density of 13.64 mA cm⁻², and a fill factor of 87.0%. All the key photovoltaic parameters of PSCs can be simultaneously increased by incorporating 1.0 vol % CN in blend solutions due to the optimized phase separation of active layers assisted by the volatilization of CN. Over 24% PCE improvement can be obtained by incorporating 1.0 vol % CN, indicating that the dynamic process of film forming should play the vital role in determining the performance of PSCs.

1. INTRODUCTION

Polymer solar cells (PSCs) have achieved rapid development during the past decade, especially for nonfullerene materials as electron acceptors.¹⁻⁶ Many efforts have been made to improve the performance of PSCs from material synthesis and device engineering sides. Tandem or ternary strategy has been commonly investigated to improve the performance of PSCs, by enhancing photon harvesting with two subactive layers or one active layer containing three different band gap materials.⁷ In fact, ternary PSCs have been demonstrated as an efficient and potential strategy due to its simple device fabrication process.⁸⁻¹¹ Nonfullerene materials have attracted more attention due to their tunable band gap, energy levels, and high absorption coefficients, leading to the enhanced open-circuit voltage (V_OC) and short circuit current density (J_SC).¹²⁻¹⁴ The dynamic process of film formation also plays the vital role in determining phase separation of active layers, which will greatly affect the exciton dissociation and charge transport efficiency. Solvent additives with different boiling point and selective dissolution of donor or acceptor have been used to adjust the phase separation for improving the performance of PSCs, such as 1,8-diiodooctane, 1-chloronaphthalene (CN), 1,8-octanediol, diphenylether, poly(dimethylsiloxane), etc.¹⁵⁻²⁰ Different solubilities of donor or acceptor in solvent additive are used to optimize the phase separation in the active layers, especially in the rapid thin-film formation process during spin-coating. Thermal annealing and solvent vapor treatment were also commonly used to optimize phase separation of the active layers. Recently, upside-down thermal annealing and upside-down solvent vapor treatment were successfully developed to finely adjust the phase separation for improving the fill factor (FF) of PSCs.²¹ The photovoltaic parameters of PSCs can be simultaneously improved by selecting appropriate materials and employing active layer treatments.

Here, the recent progress in PSCs with V_OC close to or more than 1.0 V are summarized in Table 1. Most PSCs exhibit relatively low power conversion efficiencies (PCEs) and FFs if the V_OC are close to or more than 1.0 V, which may be due to insufficient exciton dissociation of the relatively low offsets between the lowest unoccupied molecular orbitals levels of donor and acceptor. Meanwhile, there is plenty of room for FF improvement of PSCs with relatively high V_OC. In this work, polymer material PBDB-T as the donor and nonfullerene material O-IDTBR as the acceptor were selected to prepare PSCs with 1-chloronaphthalene (CN) as solvent additive. The optimized PSCs exhibit relatively high V_OC of 1.01 V and PCE of 8.09% for the active layer with 1.0 vol % CN, which are acceptable among the previous values. Compared with the PSCs without CN solvent additive, three photovoltaic parameters of PSCs can be simultaneously improved by incorporating 1.0 vol % CN, resulting from the optimized phase separation of the active layers. Figure 1 exhibits the chemical structures of the used materials, the device structure schematic diagram, and the energy levels of the used materials.
Table 1. Key Photovoltaic Parameters of PSCs with \( V_{OC} \)s Close to or More than 1.0 V

| Binary blend                  | \( J_{SC} \) (mA cm\(^{-2}\)) | \( V_{OC} \) (V) | FF (%) | PCE (%) | refs    |
|-------------------------------|-------------------------------|-----------------|-------|---------|--------|
| DR3TBDTT:O-IDTBR              | 11.06                         | 1.15            | 50    | 6.36    | 22     |
| DR3:O-IDTBR                   | 11.3                          | 1.12            | 50    | 6.1     | 23     |
| PVBDTTAZ:O-IDTBR              | 16.26                         | 1.08            | 63.6  | 11.2    | 24     |
| BDT3TR:O-IDTBR                | 12.31                         | 1.08            | 56    | 7.09    | 25     |
| PBDT-DFQX1:O-IDTBR            | 14                            | 1.07            | 57.88 | 8.67    | 26     |
| PBDB-T:ITCC                   | 15.9                          | 1.01            | 71    | 11.4    | 27     |
| PTB7-Th:O-IDTBR               | 15.7                          | 1.01            | 64.6  | 9.83    | 28     |
| PBDB-T:O-IDTBR                | 13.64                         | 1.01            | 58.7  | 8.09    | this work |
| PBDB-T:IFTN                   | 7.3                           | 1.01            | 42.1  | 3.03    | 29     |
| PBDB-T:IF-TN                  | 6.25                          | 1.01            | 41.5  | 2.57    | 29     |
| PBDB-T:IT-OM-3                | 16.38                         | 0.98            | 70    | 10.8    | 30     |
| PBTAZS-O-IDTBR                | 16.7                          | 0.98            | 63    | 10.3    | 31     |
| PBDB-T:DICTiF                 | 11.2                          | 0.98            | 65    | 7.11    | 32     |
| PBDB-T:IDT-TN                 | 11.09                         | 0.98            | 41.3  | 4.49    | 29     |
| PBDB-T:ITC6-IC                | 16.41                         | 0.97            | 73    | 11.61   | 33     |
| PBDB-T:IT-M                   | 16.82                         | 0.97            | 72    | 11.5    | 34     |
| PBDB-T:IT-DM                  | 16.48                         | 0.97            | 70.6  | 11.29   | 35     |
| PBDB-T:IT-DM                  | 16.47                         | 0.97            | 70.26 | 11.25   | 35     |
| PBDB-T:NTIC-Ome               | 13.52                         | 0.97            | 66    | 8.47    | 37     |
| PBDB-T:IDT-TN                 | 13.54                         | 0.97            | 45.64 | 5.89    | 29     |
| PBDB-T:DTNICS                 | 12.92                         | 0.96            | 72.84 | 9.03    | 38     |
| PBDB-T:CDTCN                  | 11.26                         | 0.96            | 57.58 | 6.23    | 39     |
| PBDB-T:SFTTIC                 | 9.28                          | 0.96            | 63.8  | 5.66    | 40     |
| PBDB-T:IT-M                   | 17.34                         | 0.95            | 73.2  | 12.1    | 41     |
| PBDB-T:DF-PCIC                | 13.38                         | 0.95            | 63    | 8.1     | 42     |

Figure 1. (a) Chemical structures of the used materials and schematic diagram of the device structure. (b) The energy levels of the used materials.

Figure 2. (a) Normalized absorption spectra of neat PBDB-T and O-IDTBR films. (b) Absorption spectra of blend films with different CN doping volume ratio.
2. RESULTS AND DISCUSSION

The absorption spectra of neat PBDB-T and O-IDTBR films were recorded, and the normalized absorption spectra are shown in Figure 2a. The absorption peaks of neat PBDB-T and O-IDTBR film are about 625 and 695 nm, respectively, with a large spectral overlapping from 600 to 650 nm. The absorption spectra of blend films with different CN doping volume ratio were measured and are shown in Figure 2b. The absorption spectra of blend films exhibit a marked absorption peak at about 630 nm, resulting from the large absorption spectral overlapping of the neat films. The effect of CN doping volume ratio on the absorption intensity of blend films can be mostly neglected, the slightly decreased absorption intensity may be due to the low concentration of mixed solutions with more CN. To investigate the effect of CN doping volume ratio on the performance of PSCs, the current density versus voltage (J–V) curves were measured under AM 1.5G illumination with light intensity of 100 mW cm$^{-2}$, as shown in Figure 3a.

Markedly different photovoltaic parameters can be clearly observed from the J–V curves of the corresponding PSCs for the active layers with different CN doping volume ratios, although the absorption spectra of all blend films almost overlapped. The PCE of PSCs without CN is 6.48%, with a relatively low $J_{SC}$ of 12.95 mA cm$^{-2}$, $V_{OC}$ of 0.99 V, and FF of 50.35%. For the active layers with different CN doping volume ratios, the FFs and $J_{SC}$s of the PSCs can be markedly improved and then slightly decreased along with the increase of the CN doping volume ratio. Meanwhile, the $V_{OC}$s of the PSCs can be kept almost constant, with a rather small fluctuation of less than 0.03 V. The PCE of the optimized PSCs reaches 8.09% for the active layer with 1.0 vol % CN, along with a $J_{SC}$ of 13.64 mA cm$^{-2}$, a $V_{OC}$ of 1.01 V, and a FF of 58.70%. The marked increase in FF exhibits that the phase-separation degree can be well optimized by incorporating appropriate CN in the blend solutions.

### Table 2. Key Parameters of PSCs with Different CN Doping Volume Ratios

| CN (vol %) | $J_{SC}$ (mA cm$^{-2}$) | $V_{OC}$ (V) | FF (%) | PCE (%) |
|------------|--------------------------|-------------|--------|---------|
| without    | 12.95                    | 0.99        | 50.35  | 6.48    |
| 0.5        | 13.30                    | 1.01        | 56.75  | 7.62    |
| 1.0        | 13.64                    | 1.01        | 58.70  | 8.09    |
| 1.5        | 13.17                    | 1.01        | 58.07  | 7.73    |
| 2.0        | 12.77                    | 1.02        | 57.30  | 7.46    |
| Avg.       | 13.30                    | 1.01        | 58.5   | 7.60    |

*Avg. PCE values were calculated according to 20 cells prepared from different batches.*

Figure 3. (a) $J$–$V$ curves of PSCs with different CN doping volume ratio in blend solutions. (b) External quantum efficiency (EQE) spectra of the corresponding PSCs.

Figure 4. (a) $J_{PH}$–$V_{eff}$ curves of PSCs processed without or with 1 vol % CN. (b) $J_{SC}$ dependence on light intensity of the corresponding PSCs.
resistance ($R_s$) and shunt resistance ($R_{sh}$) can be calculated according to the $J–V$ curves of PSCs without or with different CN doping volume ratios. The optimized PSCs with 1.0 vol % CN exhibits a minimal $R_s$ of 12 Ω cm$^2$ and a maximal $R_{sh}$ of 647 Ω cm$^2$, which can well support the relatively high FF. The key parameters of PSCs with different CN doping volume ratios are summarized in Table 2. The external quantum efficiency (EQE) spectra of all the PSCs were recorded and are shown in Figure 2b. The EQE values of the PSCs with 1.0 vol % CN are slightly larger than those of other PSCs, which may result from the optimized exciton dissociation, charge transport, and collection dependence on phase separation.

To further investigate the effect of CN doping volume ratio on the performance of PSCs, the $J–V$ curves of the PSCs were measured in dark and under one standard simulation solar light illumination. The photocurrent density ($J_{ph}$) can be calculated according to the equation: $J_{ph} = J_a - J_d$ where $J_a$ and $J_d$ are the current densities under 100 mW cm$^{-2}$ light illumination and in dark conditions.$^{43–45}$ The $J_{ph}$ versus effective voltage ($V_{eff} = V_a - V_s$) curves of the PSCs without or with 1.0 vol % CN are exhibited in Figure 4a, where $V_a$ is the voltage for $J_{ph} = 0$ and $V_s$ is the applied bias. It is apparent that $J_{ph}$ of the PSCs with 1.0 vol % CN rapidly reaches the saturated states compared with the others.

### Table 3. $J_{ph}$, $J_{sat}$, $G_{max}$ and $J_{ph}/J_{sat}$ Values of the PSCs Processed without or with 1 vol % CN

| CN (vol %) | $J_{ph}^a$ (mA cm$^{-2}$) | $J_{ph}^b$ (mA cm$^{-2}$) | $J_{sat}$ (mA cm$^{-2}$) | $J_{ph}/J_{sat}^a$ (%) | $J_{ph}/J_{sat}^b$ (%) |
|-----------|--------------------------|--------------------------|--------------------------|------------------------|------------------------|
| without   | 13.17                    | 9.72                     | 14.91                    | 88.3                   | 65.2                   |
| 1.0       | 13.40                    | 10.99                    | 14.51                    | 92.3                   | 75.8                   |

$^a$Short-circuit condition. $^b$Maximal power output condition.

PSCs without CN under a relatively low bias, indicating the more efficient transport charge in the active layer with 1.0 vol % CN. According to the $J_{ph}$ versus $V_{eff}$ curves, the exciton dissociation and the charge collection efficiency can be evaluated by the ratios of $J_{ph}$ to saturation photocurrent density ($J_{sat}$) at short circuit or maximal power output conditions, respectively. The detailed values of the PSCs without and with 1.0 vol % CN are summarized in Table 3. The exciton dissociation and charge collection efficiency can be simultaneously improved by adding 1.0 vol % CN, resulting from the optimized phase separation. To further investigate the positive effect of 1.0 vol % CN on the performance of PSCs, the $J–V$ curves of PSCs without and with 1.0 vol % CN were measured under different light intensities ($P_{light}$) from 1 to 100 mW cm$^{-2}$. According to the $J–V$ curves, the relationship between $J_{sc}$ and $P_{light}$ are presented in Figure 4b, which can be evaluated by the power-law formula of $J_{sc} \propto P_{light}^{α}$. The bimolecular recombination in active layers can be negligible if the fitted parameter $α$ is close to 1. The fitted parameter $α$ is about 0.853 or 0.866 for the PSCs without or with 1.0 vol % CN, which are slightly far away from 1. The bimolecular recombination in both kinds of active layers cannot be neglected, resulting in the relatively low FF of PSCs. The FF of 58.70% for PSCs with 1.0 vol % CN is slightly larger than that of 50.35% for PSCs without CN, which can be well explained according to the values of the fitted parameter $α$.

The hole-only and electron-only devices were fabricated to investigate the effect of CN doping volume ratio on the charge mobility in the active layers with the configuration of indium tin oxide (ITO)/poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS)/active layers/MoO$_3$/Ag and ITO/ZnO/active layers/2,9-bis(3-(dimethylamino)propyl)anthra[2,1,9-def:6,5,10-def′′]diisoquinoline-1,3,8,10(2H,9H) tetraone (PDIN)/Al, respectively. The $\ln(Jd/V^2)−(V/d)^{0.5}$ curves of hole-only and electron-only devices are shown in Figure 5a,b. The electron mobility ($\mu_e$) and hole mobility ($\mu_h$) can be calculated according to the $\ln(Jd/V^2)−(V/d)^{0.5}$ curves, as listed in Table 4. It is apparent that the hole and electron mobilities in the active layers can be increased and decreased along with the increase in CN doping volume ratio. For the active layers with 1.0 vol % CN, $\mu_e$ and $\mu_h$ reach 3.93 $\times$ 10$^{-4}$ and 7.10 $\times$ 10$^{-4}$ cm$^2$ V$^{-1}$ s$^{-1}$, respectively. Meanwhile, the hole and electron transport in the active layers with 1.0 vol % CN become more balanced, which is beneficial to improve the charge collection in the corresponding PSCs. The ratio of $\mu_h$ to $\mu_e$ reaches 1.81 for the PSCs with 1.0 vol % CN, leading to the relatively large FF of PSCs. As we known, both photogenerated
3. CONCLUSIONS

A series of PSCs with PBDB-T as donor and O-IDTBR as acceptor were prepared with different CN doping volume ratios to adjust the phase separation degree of the active layers. The optimized PSCs exhibit a PCE of 8.09%, with a $J_{SC}$ of 13.64 mA cm$^{-2}$, a $V_{OC}$ of 1.01 V, and a FF of 58.70% for the active layers with 1.0 vol% CN. Compared with the PSCs without CN, all of the photovoltaic parameters can be simultaneously improved by incorporating appropriate CN to optimize the phase separation. More than 24% PCE improvement can be obtained by incorporating 1.0 vol% CN into the blend solutions, indicating that the dynamic process of thin film formation should play a vital role in determining the performance of PSCs.

4. EXPERIMENTAL SECTION

The indium tin oxide (ITO) coated glass substrates ($15 \Omega \text{sq}^{-1}$) were continuously cleaned by sonication in detergent, deionized water, and ethanol. The cleaned ITO substrates were dried by high-purity nitrogen and treated by oxygen plasma for a minute to further improve their working function and clearance. Afterward, poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate) (PEDOT:PSS, purchased from H.C. Starck co. Ltd.), as a hole transport layer, was spin-coated on the ITO substrates at 5000 rpm for 40 s and dried at 150 °C for 10 min in atmospheric air. Then, ITO substrates coated with PEDOT:PSS films were transferred into a high-purity nitrogen-filled glovebox to fabricate active layers. The used materials, PBDB-T and O-IDTBR, were purchased from Organicsemiconductor Materials Inc. and Derthon Optoelectronic Materials Science Technology Co LTD, respectively. The mixed PBDB-T/O-IDTBR powder with a weight ratio of 1:1 was dissolved in chlorobenzene to prepare 20 mg/mL blend solutions and different 1-chloronaphthalene (CN) volumes (0.5, 1.0, 1.5, and 2.0 vol%) were doped into the blend solutions. The solutions were spin-coated onto the PEDOT:PSS/ITO substrates at 2300 rpm for 40 s to prepare the active layers. And then, the prepared active layers were annealed at 80° for 10 min in a high-purity nitrogen-filled glovebox. The small molecule 2,9-bis-(3-(dimethylamino)propyl)anthra-[2,1,9-def:6,5,10-d’e’f’]disoquinoline-1,3,8,10(2H,9H) tetrone (PDIN) was dissolved in methanol with the addition of 0.28 vol% acetic acid to prepare a 2 mg/mL solution. Then, the prepared PDIN solution was spin-coated onto the active layers at 3000 rpm for 40 s to prepare the cathode interfacial layer. Finally, 100 nm aluminum (Al) was deposited with a mask by thermal evaporation under vacuum of $4 \times 10^{-3}$ Pa. The active area is approximately 3.8 mm$^2$, which is defined by the overlapping area of ITO anode and Al cathode. A series of PSCs were fabricated with the configuration ITO/PEDOT:PSS/active layer/PDIN/Al, with the only difference of the CN doping volume ratios in the blend solutions. The detailed characteristics of the PSCs and the corresponding blend films are the same with that reported in our previous works.

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ACKNOWLEDGMENTS

This work was supported by the National Training Program of Innovation and Entrepreneurship for Undergraduates (Grant No. 180170006), National Natural Science Foundation of China (61377029, 61564003, 61675017, 21572171, and 61705161).

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