Research Article

Investigation on Thermal Degradation Process of Polymer Solar Cells Based on Blend of PBDTTT-C and PC₇₀BM

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The effects of thermal treatment on the photovoltaic performance of conventional and inverted polymer solar cells (PSCs) based on the combination of poly[(4,8-bis-(2-ethylhexyloxy)-benzo[1,2-b:4,5-b']dithiophene)-2,6-diyl-alt-(4-(2-ethylhexanoyl)-thio-no[3,4-b]thiophene))-2,6-diyl] (PBDTTT-C) and [6,6]-phenyl C₇₀-butyric acid methyl ester (PC₇₀BM) are investigated. The transient photoconductivity, the absorption spectra, and the transmission electron microscopy (TEM) images have been employed to study the thermal degradation of the inverted PSCs. The degradation is attributed to the inefficient charge generation and imbalance in charge-carrier transport, which is closely associated with the morphological evolution of the active layer with prolonged heating time.

1. Introduction

Bulk heterojunction (BHJ) PSCs have been paid much attention as candidates for sustainable solar energy converters because of their unique advantages of low cost, light weight, large area, and simple solution-based fabrication with mechanical flexibility. The power conversion efficiency (PCE) has been extensively enhanced using low-bandgap materials, optimizing the device structures and controlling the morphology of active layers [1–4]. Although the reported PCEs nearly meet the initial requirements for commercial applications, the poor stability of the PSCs is still a major barrier to realize the commercialization [5–8]. Improvements are necessary to enhance the stability of these devices.

Related studies have suggested decay mechanisms, such as the electrode degradation under the presence of oxygen and moisture [9], metal ion diffusion into the active layers from the electrodes [10], and photoactive material degradation with oxygen and moisture upon illumination and

heating of the device [11–14]. Several groups have investigated the thermal degradation of PSCs by simulating the actual working environment under sunlight. For instance, Wong et al. reported that the microscopic and nanoscopic crystallization of [6,6]-phenyl C₆₀-butyric acid methyl ester (PC₆₀BM) influenced the morphological stability and performance of polymer-fullerene solar cells under thermal stress [15].

However, the mechanisms of PSC degradation under thermal stress, as well as the sustained morphological changes and charge transport kinetics, remain unclear. In this study, inverted and conventional PSCs based on the blend of PBDTTT-C and PC₇₀BM were fabricated. The thermal stability and optoelectronic behavior of these devices were investigated via morphological analyses and transient photocurrent measurements.

2. Experimental

2.1. Materials. ZnO precursor was prepared by dissolving 0.3 mol zinc acetate dehydrate (Zn (OOCCH₃)₂·2H₂O, ACS,

FIGURE 1: (a) Molecular structures of PBDTTT-C and PC₇₀BM. (b) The structure of the inverted polymer solar cell. (c) The structure of the conventional polymer solar cell.

98.0–101.0%) in 0.3 mol ethanolamine (HOCH₂CH₂NH₂, ACS, 99+ %) and 2-methoxyethanol (C₃H₈O₂, ACS, 99.3%). PBDTTT-C (molecular weight: >20,000 g/mol and PDI: 1.8–2.5) was purchased from Solarmer Materials Inc, and PC₇₀BM was purchased from Solenne b.v. The corresponding chemical structures are depicted in Figure 1(a). 1, 8-Diiodooctane (DIO) was purchased from Alfa Aesar. Poly (3, 4-ethylenedioxythiophene):poly (styrenesulphonic acid) (PEDOT : PSS) (Clevious P VP AI 4083) was purchased from H. C. Starck Company.

2.2. Fabrication of Solar Cells. Inverted PSCs were fabricated with the structure of glass/ITO/ZnO/PBDTTT-C:PC₇₀BM/MoO₃/Ag (Figure 1(b)). ITO glasses were cleaned with detergent, deionized water, acetone, and isopropanol and subsequently dried with N₂ flow. ZnO films were prepared by spin coating from the ZnO precursor

solution at a speed of 4000 rpm for 50 s and baked at 150°C for 5 min at ambient air. Then the organic residues in the ZnO film were removed by ultrasonic cleaning in deionized water, acetone, and isopropyl alcohol. The active layer was then prepared by spin coating from dichlorobenzene solution of PBDTTT-C and PC₇₀BM (1:1.5 w/w, polymer concentration of 10 mg/mL) with 3% volume ratio of DIO additive on ZnO films with thickness of ~80 nm in a nitrogen-filled glove box. Finally, 15 nm MoO₃ and 100 nm Ag were thermally deposited on the active layer under the pressure of 4×10^{-4} Pa. The active area of the device was 4.5 mm².

Conventional PSCs were fabricated with the structure of glass/ITO/PEDOT:PSS/PBDTTT-C:PC₇₀BM/Ca/Al (Figure 1(c)). The precleaned ITO substrates were treated with UV-ozone for 20 min, then modified by spin-coating PEDOT:PSS with a thickness of around 40 nm, and then baked at 150°C for 15 minutes at ambient conditions. Then

FIGURE 2: DSC thermograms showing the heating processes for the pure PC₇₀BM (a), pure PBDTTT-C (b), and PBDTTT-C/PC₇₀BM blends (c).

TABLE 1: Device parameters of the conventional and inverted devices which are kept at 70°C for different time.

Device structures	Time (h)	Jsc (mA/cm ²)	Voc (V)	FF (%)	PCE (%)
Conventional	Control	15.41	0.71	60.56	6.62
	1	14.91	0.71	58.34	6.17
	2	14.79	0.70	58.99	6.11
	3	14.77	0.71	57.94	6.07
	5	14.37	0.71	58.97	6.01
	18	13.71	0.72	58.38	5.76
Inverted	Control	15.28	0.72	61.33	6.74
	1	15.12	0.72	61.40	6.68
	2	14.66	0.71	62.64	6.52
	3	14.64	0.71	62.37	6.48
	5	14.59	0.71	62.46	6.47
	18	13.86	0.71	62.08	6.11

FIGURE 3: J - V curves of the inverted solar cells treated at different temperatures.

the substrates with PEDOT:PSS were transferred to glove box to fabricate the photoactive layer at experiment condition similar to those in the inverted device. Finally, 20 nm Ca and 100 nm Al were thermally deposited on the active layer as cathodes.

2.3. Device Characterizations. The current density-voltage (J - V) measurements of the devices were performed by a computer-controlled Keithley 6430 Source Measure Unit. Photovoltaic characterization was conducted under simulated AM1.5G irradiation (100 mW/cm^2) of solar simulator XES-301S+EL-100 (SAN-EI ELECTRIC). The thermal-stability tests were carried out on a hotplate (IKA RCT basic magnetic stirrer) as heating source to control temperature at an accuracy of $\pm 1^\circ\text{C}$. All electrical measurements were performed in a nitrogen-filled glove box. A UV/visible spectrophotometer (Shimadzu UV-3101) was used to characterize the absorption properties of the PBDTTT-C:PC₇₀BM layer. TEM images were obtained by using a Tecnai G2 F30 transmission electron microscope operated at 300 kV.

PBDTTT-C and PC₇₀BM spin-coated on glass substrate were collected for differential scanning calorimetry (DSC) measurements, which were performed on a NETZSCH DSC 200F3 equipment. N₂ (flux about 60 mL/min) was used as the purge gas. Approximately 5 mg of the samples was sealed in perforated aluminum crucibles. The scan rate during DSC measurements was set to 5 K/min.

To measure transient photoconductivity, the samples were excited by an optical parametric oscillator (OPO) pumped by a Q-switched neodymium ion-doped yttrium aluminum garnet (Nd³⁺:YAG) laser. The OPO delivered pulses with a duration of about 5 ns, a wavelength of 460 nm, and an energy of 3 nJ per pulse at a repetition rate of 10 Hz. The carriers of the devices were generated by a short laser pulse; then the transient photocurrent was obtained by

measuring the voltage drop across a 5Ω resistor load that was connected in series with the solar cell; the current traces were recorded by a Tektronix TDS 540D oscilloscope.

3. Results and Discussion

High temperatures can accelerate PSCs degradation [16]. The Arrhenius model describes this process [17], in which the degradation constant is defined as follows:

$$k_{\text{deg}} = A \exp\left(\frac{-E_a}{k_B T}\right), \quad (1)$$

where E_a is the activation energy, k_B is the Boltzmann constant, T is the temperature (K), and A is a constant dependent on the degradation mechanisms and the experimental conditions. Typically, degradation process is in close association with the intrinsic properties of materials, such as the trapping behavior of electrical carriers and structural and chemical changes of polymers. The activation energy E_a is a common parameter that the activated processes have to overcome. Hence, the kinetic processes are strongly temperature dependent.

Hou et al. reported that the glass transition temperature of PBDTTT-C is about 400°C to 410°C , which is above the operating temperature of the device [18]. In this study, the thermal behavior of PBDTTT-C, PC₇₀BM, and their blends is investigated by DSC to determine the temperature of phase transitions. The traces of heat flow during the annealing of PBDTTT-C, PC₇₀BM, and their blends are illustrated in Figure 2. For pure PC₇₀BM, an exothermic peak (197°C) and a melting peak (204°C) are observed in Figure 2(a). From Figure 2(c), we can find a new distinguishable peak locating at 64.3°C defined as the eutectic temperature of the blend of PBDTTT-C and PC₇₀BM. The new peak corresponds to the melting temperature of bimolecular crystals formed by fullerene intercalation between the polymer side chains [19, 20]. The well-ordered bimolecular crystals can cause strong charge-transfer states due to the strong coupling of the dissimilar nearest neighbor molecules [20, 21]. So the reduction of bimolecular crystals would affect the electronic properties of the PSCs such as exciton dissociation, recombination, and charge transport.

Considering our DSC results and the actual working temperature of the solar cells in outdoor environments (in the range of 30°C to 80°C), the temperature ranging from 50°C to 90°C was selected to investigate PSCs degradation. All measurements were performed in a glove box to avoid oxygen and water induced degradation. Figure 3 reveals that the performance of devices exhibits burn-in losses under thermal stress at different temperatures for 1 h. The open-circuit voltage (V_{oc}) is stable at 0.72 V , and the short-circuit current density (J_{sc}) acts as the major degradation factor. The heated devices display a slightly decline in J_{sc} compared with the control device when the temperature is below 70°C . However, J_{sc} shows a steep decrease at 80°C and 90°C . The decrease of the J_{sc} means the reduced interfacial area between the donor polymer and the acceptor fullerene after the annealing [22]. Integrating the results of DSC measurements, we selected

FIGURE 4: J - V curves of the (a) conventional and (b) inverted solar cells treated at 70°C for different time.

FIGURE 5: Normalized PCE, FF, Voc, and Jsc of conventional (red dot) and inverted (black square) solar cells at 70°C for different time. All curves are normalized to the initial value.

70°C as the actual condition temperature to analyze the effects of heating duration on the long-term performances of PSCs.

The J - V characteristics of PSCs are shown in Figure 4, and the performance of the devices is summarized in Table 1. The conventional and inverted devices exhibit significantly reduced performances after heat treatment at 70°C for 18 h. For conventional devices, efficiency decreases from 6.62% to 5.76%, Jsc decreases from 15.29 mA/cm^2 to 14.76 mA/cm^2 , and fill factor (FF) drops from 60.56% to 58.28%. For inverted devices, Jsc decreases from 15.28 mA/cm^2 to 13.86 mA/cm^2 , and FF increases to a maximum value of 62.64% after 2 h heating treatment and then slightly decreases to 62.08%. Variations in the parameters result in PCE degradation from 6.74% to 6.11%. To further compare the photovoltaic performances of conventional and inverted PSCs, the normalized PCE, FF, Voc, and Jsc are shown in Figure 5. Jsc of the conventional devices is degraded by 11%, which is slightly higher than that of the inverted devices (9%). Although Voc and FF of both PSCs fluctuate during heating, inverted PSCs exhibit a more stable FF value. PCE of the inverted PSCs decreases by 10%, whereas that of the conventional devices drops to 85% of the original PCE during 18 h thermal treatment. The better thermal stability of the inverted PSCs may be attributed to the natural self-encapsulated structure that comprised a well vertical phase separation in the active layer [23, 24], on which the metal oxides (MoO_3) and stable metals (Ag) are deposited as the buffer layer and top electrode, respectively [25, 26]. Considering that the inverted PSCs have better thermal stability than that of the conventional PSCs, we choose the inverted PSCs as the study object in the following discussion.

In general, Jsc is determined by the generation, transport, and extraction of charge carriers. Reduction in this parameter is larger than that of the other parameters during the

FIGURE 6: (a) Decay of the transient photocurrent density J and normalized J (insert) of control device and the devices thermally treated at 70°C for different time. (b) Total swept-out charge density as a function of decay time for solar cells.

FIGURE 7: Absorption spectra of the (a) inverted solar cells and the normalized absorption spectra of (b) inverted solar cells treated at 70°C for different time.

prolonged heating. The transient photocurrent is measured to investigate the influence of thermal treatment on charge transport kinetics [27]. The transient photoconductivity of the inverted devices treated with different heating times is shown in Figure 6. As shown in the inset of Figure 6(a), the transient photocurrent curves of devices reflect the rates of the extracted carriers in the BHJ active layer, which could result in the change of the FF values. The temporal dependence of the total swept-out charge density calculated by integrating the photocurrent decay is shown in Figure 6(b). The total charges of the PSCs after thermal

treatment significantly decreased compared with those of the control devices. The number of mobile carriers constantly decreases with increasing heating time.

The extraction of photogeneration charges is low because of the reduced absorption with prolonged heating time [28]. To further study the photogeneration charges of the PSCs after thermal treatment, the corresponding absorption spectra were measured (Figure 7). The absorption intensity of inverted PSCs decreases with increasing heating time because of the unstable morphological evolution (Figure 7(a)). This evolution leads to the excessive phase separation in the

FIGURE 8: TEM images of the PBDTTT-C: PC₇₀BM films treated at 70°C for control (a), 1 hour (b), 2 hours (c), 3 hours (d), 5 hours (e), and 18 hours (f).

blend films. The absorption of PSCs increases from 650 nm to 670 nm with the heating time after thermal treatment based on the normalized absorption spectra (Figure 7(b)). The absorption maximum exhibits a redshift with increasing heating time, which indicates that the high crystalline order and large domain sizes are obtained during the thermal process. The charge-carrier transport is unbalanced because of the uneven crystalline state (Figure 6) [29, 30].

TEM images of the blend films which are obtained before and after thermal treatment are given in Figure 8. The as-casted blend film exhibits a well-developed phase separation with the PBDTTT-C and the PC₇₀BM uniformly distributed across the network of the blend film and donor-acceptor

domains with appropriate sizes (Figure 8(a)). The outlines of the phase separation between PBDTTT-C and PC₇₀BM are very clear as shown in Figure 8(a). The interpenetrating networks with appropriate phase sizes can provide a favorable interface for exciton dissociation and pathways for charge-carrier transport without too many chances of recombination during the transit processes [16, 22]. The distribution of the donor-acceptor phases becomes more compact, when the heating time is increased from 1 h to 5 h (Figure 8(b) to Figure 8(e)). It means that the clusters of the PC₇₀BM have grown with big sizes by the incorporating molecules drawn from the distributed PC₇₀BM network. Subsequently, the states of the domains become ambiguous. With the growing

numbers and sizes, the clusters touch each other and deplete the polymer matrix due to lack of space. The outlines of the phase separation between the PC₇₀BM and PBDTTT-C appear to be blurry. The TEM image of the blend film reveals the phase separation with large domain size after 18 h thermal treatment, which changes the absorption, exciton dissociation, and carriers transport.

4. Conclusion

In summary, the effects of heating temperature and heating time on the thermal stability of conventional and inverted PSCs based on PBDTTT-C:PC₇₀BM in a N₂ atmosphere are analyzed. The results reveal that the inverted PSCs exhibit better thermal stability than the conventional devices. The absorption of the blend film decreases with increasing heating time because of the formation of high crystalline orders and large domain sizes. Transient photoconductivity results indicate that the inefficient charge generation and nonequilibrium charge-carrier transport may result in the thermal degradation of PSCs. These results provide methods to enhance the thermal stability of the PSCs.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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