Influence of Physical Load on the Stability of Organic Solar Cells with Polymer : Fullerene Bulk Heterojunction Nanolayers

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ABSTRACT: We report the effect of physical load on the stability of organic solar cells under physical loads. The active layers in organic solar cells were fabricated with bulk heterojunction films (BHJ) films of poly (3-hexylthiophene) and phenyl- C_{61} -butyric methyl ester. The loading time was varied up to 60 s by keeping the physical load constant. Results showed that the open circuit voltage was not influenced by the physical load but other solar cell parameters were sensitive to the loading time. The fill factor was very slightly increased at 15 s, while short circuit current density was well kept for 30 s. The power conversion efficiency was reasonably maintained for 45 s but became significantly decreased by the continuous loading for 60 s.

Key words: Organic solar cells, Bulk heterojunction layer, Physical loading, Stability, Power conversion efficiency

1. Introduction

Organic solar cells have been spotlighted as one of the most viable next generation solar cells due to their potentials for ultrathin and lightweight low-cost solar modules¹⁻⁶⁾. Of various organic solar cells, polymer:fullerene solar cells have been extensively studied because of high power conversion efficiency (PCE) approaching ~12% via gradual advances in light-absorbing semiconducting polymers and fullerene derivatives⁷⁻¹³⁾. In addition, polymer:fullerene solar cells can be manufactured by employing various wet-coating/printing technologies with continuous roll-to-roll processes at low temperatures, which may enable custom-designed plastic solar modules leading to wide spreading of solar cells in our daily life³⁾.

In principle, the light-absorbing layers in polymer:fullerene solar cells consist of bulk heterojunction (BHJ) structures that are made between conjugated polymer chains and fullerene derivative aggregates. The resulting BHJ layers should have a proper charge percolation path for the efficient transport of individual charges (electrons and holes) separated from the excitons generated by light absorption. Thus the nanoscale morphology of BHJ layers plays a critical role in achieving such high

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PCE¹⁴⁻¹⁶⁾. On this account, a variety of nanomorphology studies have been carried out to optimize the charge percolation paths according to various kinds of conjugated polymers for polymer: fullerene solar cells¹⁷⁻¹⁹⁾.

As a result, a scenario has been built to achieve reasonably high PCEs for different kinds of polymer materials. A thermal annealing method has been proposed for crystalline polymerbased BHJ systems²⁰⁻³⁶, while an additive method is currently used for less-crystalline (amorphous) polymer-based BHJ layers³⁷⁻⁴⁷. However, it is doubtable that such an optimized BHJ layer (system) can withstand external physical loads such as pressing, hitting, rubbing and the like when it comes to the fullerene nano-domains dispersed in the polymer:fullerene mixture structures.

In this work, we have investigated the stability of polymer: fullerene solar cells under a physical load (11.3 kg/cm^2) according to the loading time. To get an insight for the physical loadinduced stability change, a well-established polymer:fullerene solar cell with the BHJ layer of poly (3-hexylthiophene) (P3HT) and [6,6]-phenyl-C₆₁-butyric acid methyl ester (PC₆₁BM) has been first introduced.

2. Experimental Section

2.1 Materials and solutions

The P3HT polymer (regionegularity = 95%, weight-average

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molecular weight = 53 kDa, polydispersity index = 1.5) was provided from Solaris Chem Inc. (Canada), while $PC_{61}BM$ (purity = 99.5%, formula weight = 910.9 Da) was used as received from Nano-C (USA). The binary solutions of P3HT (25 mg) and $PC_{61}BM$ (25mg) were prepared using chlorobenzene as a solvent at a solid concentration of 50 mg/ml. For making a hole-collecting buffer layer in P3HT:PC_{61}BM solar cells, aqueous solutions of poly(3,4-ethylenedioxythiophene):poly (styrenesulfonate) (PEDOT:PSS, PH500, conductivity = 300 S/cm) were purchased from HC Starck (Clevios).

2.2 Device fabrication

To fabricate polymer:fullerene solar cells, indium-tin oxide (ITO)-coated glass substrates (sheet resistance = $10 \Omega/\Box$) were first patterned to make the ITO electrodes (8 mm × 12 mm) via photolithography/etching processes. The patterned ITO-glass substrates were cleaned using acetone and isopropyl alcohol in order to remove any remained organic residues and/or particles during the photolithography/etching process. After drying the patterned ITO-glass substrates with nitrogen flows, the surface of the dried ITO-glass substrates was treated with a UV-ozone cleaner. Then the PEDOT:PSS layer (thickness = 40 nm), as a hole-collecting buffer layer, was spin-coated on the UV-ozonetreated ITO-glass substrates, followed by thermal annealing at 230 °C for 15 min. After cooling the PEDOT:PSS-coated ITOglass substrates, the BHJ (P3HT:PC₆₁BM) layers were spin-coated on the PEDOT:PSS layer parts of the ITO-glass substrates. The BHJ layer-coated samples were soft-baked at 60 °C for 15 min and loaded into a vacuum chamber inside an argon-filled glove box. Finally, aluminum (Al) electrodes (thickness = 95 nm) were deposited on the BHJ layers through a metal shadow mask. The fabricated solar cells were subject to thermal annealing at 140 °C for 30 min.

2.3 Measurements

A specialized sample holder system with a pressing part was made for the measurement of solar cell performances under a physical load (11.3 kg/cm²). The sample holder was charged with nitrogen gas during measurement. The current density – voltage (J-V) characteristics were measured using an electrometer (Keithley 2400), while the performance of solar cells was measured under illumination with simulated solar light by employing a home-built measurement system equipped with a solar simulator (92250A-1000, Newport Corp., air mass 1.5G, 100 mW/cm²). The surface of the film and top electrodes was

examined using an optical microscope (SV-55, SOMETECH).

3. Results and Discussion

As shown in Fig. 1(a), the physical load (pressure) exerted on the Al electrode part of the P3HT:PC₆₁BM solar cells, whereas the simulated solar light was illuminated through the ITO-glass substrates. The pressing part contacting the Al electrodes was finely processed with very low surface roughness so that it was found not to hurt the surface of the Al electrodes. Although the physical load is applied to the Al electrode part, the P3HT: PC₆₁BM solar cells are expected to be operated according to the charge separation/transport mechanism based on the energy band diagram in Fig. 1(b).

As shown in Fig. 2(a), the light J-V curves of devices were very slightly changed when the loading (pressing) time increased up to 45 s. However, a noticeable change in the light J-V curves was observed at 60 s. The short circuit current density (J_{SC}) was reduced from 8.62 mA/cm² to 7.96 mA/cm² after loading for 60 s, whereas the open circuit voltage (V_{OC}) was almost unchanged

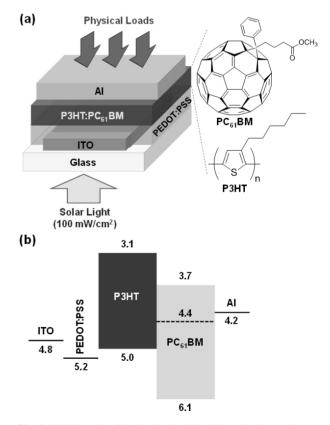


Fig. 1. (a) Illustration for the physical load (pressing) experiment of the P3HT:PC₆₁BM solar cell. (b) Flat energy band diagram for the P3HT:PC₆₁BM solar cell. Note that the minus (-) sign and energy unit (eV) in the energy band diagram were omitted in (b) (see Fig. 2(b)). Interestingly, it is found that the light J-V curve was very marginally improved by loading for 15 s as observed from the enlarged J-V curves (see inset in Fig. 2(b)).

Looking into the dark J-V curves in Fig. 3, the current density in the backward (reverse) direction (<0 V) was almost unaffected by the load irrespective of the loading time. However, the current density in the forward direction (>0 V) was gradually changed (reduced) with the loading time. When the loading time

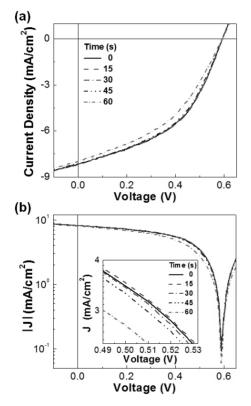


Fig. 2. Light J-V curves for the P3HT:PC₆₁BM solar cells according to the loading (pressing) time: (a) Linear scale, (b) semilogarithmic scale (inset show the enlarged J-V curves)

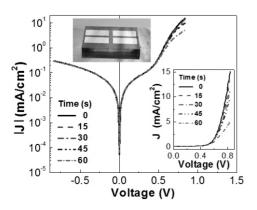


Fig. 3. Dark J-V curves on a semi-logarithmic scale (inset: linear scale) for the P3HT:PC₆₁BM solar cells according to the loading (pressing) time. The inset photograph shows the top Al electrode part of the solar cell after loading experiment

reached 30 s, the reduced current density was obviously observed at the voltage range of >0.5 V (see the linear J-V curves in the inset figure). This result indicates that the charge transport in the BHJ (P3HT:PC₆₁BM) layers is influenced by the physical load, even though the surface of the Al electrodes was almost not affected by continuous loading (pressing) of 11.3 kg/cm².

The detailed trend of solar cell parameters is plotted as a function of the loading time in Fig. 4. As discussed in Fig. 2, J_{SC} was well maintained up to 30 s, followed by gradual reduction when the loading time was extended to ≥ 45 s. However, V_{OC} was nearly insensitive to the loading time, which informs that the main bulk heterojunction morphology between P3HT and PC₆₁BM domains was excellently preserved. In contrast, the FF trend was different from the J_{SC} trend because it was marginally increased at 15 s, which can be ascribed to the slightly improved J-V curve at 15 s as shown in Fig. 2(b) inset. However, FF was

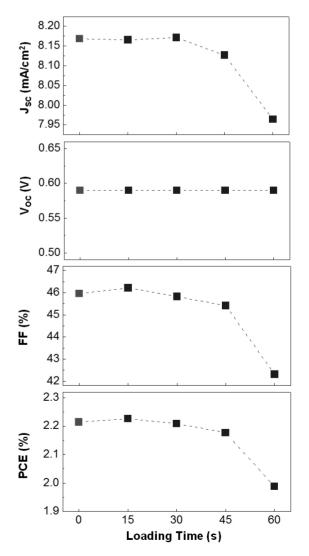


Fig. 4. J_{SC}, V_{OC}, FF and PCE as a function of the loading (pressing) time for the P3HT:PC₆₁BM solar cells

 Table 1. Summary of solar cell parameters according to the loading (pressing) time for the P3HT:PC₆₁BM solar cells

	Loading Time (s)				
	0	15	30	45	60
V _{oc} (V)	0.59	0.59	0.59	0.59	0.59
J _{SC} (mA/cm ²)	8.169	8.166	8.171	8.127	7.964
FF (%)	46.0	46.2	45.8	45.4	42.3
PCE (%)	2.215	2.226	2.209	2.178	1.988
$R_{s} (k\Omega \cdot cm^{2})$	0.26	0.26	0.27	0.28	0.34
R_{SH} (k $\Omega \cdot cm^2$)	1.9	2.3	2.7	5.1	1.4

initialized to decrease even at 30 s though J_{SC} was well kept at this time. This result may indicate that the charge transport in the BHJ (P3HT:PC₆₁BM) layers was already slightly affected by the physical load prior to its final influence on J_{SC}. Further increase in the loading time resulted in the pronounced reduction in FF, which is in agreement with the J_{SC} trend. Consequently, the power conversion efficiency (PCE) was very slightly increased up to PCE = 2.23 % at 15 s but showed gradual decreasing trend with time by further loading. As summarized in Table 1, the series resistance (Rs) was similarly starting to increase at 30 s even though it was maintained initially at 15 s. However, the shunt resistance (R_{SH}) was gradually increased up to 5.1 k Ω ·cm² at 45 s and then abruptly reduced to 1.4 k Ω ·cm² at 60 s. Thus it is expected that the leakage paths in the BHJ (P3HT:PC₆₁BM) layers might be considerably increased by loading with 11.3 kg/cm^2 for 60 s.

4. Conclusions

The stability of polymer:fullerene (P3HT:PC₆₁BM) solar cells under a physical load (11.3 kg/cm²) was investigated by varying the loading time up to 60 s. The light J-V curves were subject to marginal change upon the physical load when the loading time was 30 s. However, the shape of J-V curves was pronouncedly changed by the physical loading for 60 s, as supported by the gradually reduced dark current density with the loading time. The detailed analysis for the solar cell parameters revealed that the FF value was slightly increased by the physical loading for 15 s in the presence of the well-kept J_{SC} up to 30 s as well as no change in V_{oc}. As a result, the device efficiency (PCE) was marginally increased at 15 s, whereas further loading for 60 s led to the noticeable reduction in PCE. We expect that the present result may contribute to the stability study of organic solar cells under physical loading and/or touch that is considered another issue encountered for commercialization of organic solar cells.

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