Long-Term Shelf Lifetime of Polymer:Nonfullerene Solar Cells Stored under Dark and Indoor Light Environment

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ABSTRACT: Here we report the long-term stability of polymer:nonfullerene solar cells which were stored under dark and indoor light condition. The polymer:nonfullerene solar cells were fabricated using bulk heterojunction (BHJ) layers of poly[(2,6-(4,8-bis(5-(2-ethylhexyl) thiophen-2-yl)-benzo[1,2-b:4,5-b′]dithiophene))-alt-(5,5-(1′,3′-di-2-thienyl −5′,7-bis(2-ethylhexyl)benzo[1′,2′-c:4′,5′-c′]dithiophene-4,8-dione))] (PBDB-T) and 3,9-bis(6-methyl-2-methylene-(3-(1,1-dicyanomethylene)-indanone))-5,5,11,11-tetrakis(4-hexylphenyl)-dithieno[2,3-d:2′,3-d′]-s-indaceno[1,2-b:5,6-b′]dithiophene (IT-M). To investigate their long-term stability, the PBDB-T:IT-M solar cells were stored in an argon-filled glove box. One set of the fabricated solar cells was completely covered with an aluminum foil to prevent any effect of light, whereas another set was exposed to indoor light. The solar cells were subjected to a regular performance measurement for 40 weeks. Results revealed that the PBDB-T:IT-M solar cells underwent a gradual decay in performance irrespective of the storage condition. However, the PBDB-T:IT-M solar cells stored under indoor light condition exhibited relatively lower power conversion efficiency (PCE) than those stored under the dark. The inferior stability of the solar cells under indoor light was explained by the noticeably changed optical absorption spectra and dark spot generation, indicative of degradations in the BHJ layers.

Key words: Long-term stability, Polymer:nonfullerene solar cells, Shelf lifetime, PBDB-T, IT-M, Indoor light

1. Introduction

Organic solar cells have gained attention owing to their advantages, such as vacuum-free processes[1-5] for flexible solar modules at room temperature and low manufacturing cost[6-10]. The power conversion efficiency (PCE) of organic solar cells fabricated using bulk heterojunction (BHJ) layers has been improved up to >16% using new device structures[11-14] and novel organic materials[15-19]. In particular, an inverted-type device structure played a crucial role in the enhanced PCE of organic solar cells with the BHJ layers based on conjugated polymers as electron-donating materials[20-25].

In high-efficiency organic solar cells, fullerene derivatives as electron acceptors in BHJ layers were replaced by nonfullerene materials to improve their poor performance and stability to overcome their easy crystallization[26-30]. Compared with fullerene derivatives, nonfullerene materials have structural advantages, including their contribution to light absorption up to the near-infrared (NIR) region and excellent charge transport owing to their planar core group with more than five heterocycles[31-33].

Most results of the investigation on polymer:nonfullerene solar cells have focused on improving the PCE based on the structural tuning of the side chain and a number of heterocycles in nonfullerene materials and/or composition ratio between polymers (electron donors) and nonfullerene materials (electron acceptors)[34-37]. In this regard, the stability of polymer:nonfullerene solar cells has rarely been considered, although it is a crucial criterion to approach commercialization. Therefore, the stability investigation for long-term storage of polymer:nonfullerene solar cells under the dark and indoor light conditions is vital to confirm the effect for prolonged use of commercial solar cell products.

In this work, we tried to investigate the long-term stability of inverted-type polymer:nonfullerene solar cells which were stored in the dark and under indoor light condition. The inverted-type polymer:nonfullerene solar cells were fabricated by employing the BHJ layers that were composed of poly[(2,6-(4,8-bis(5-(2-ethylhexyl)thiophen-2-yl)-benzo[1,2-b:4,5-b′]dithiophene))-alt-(5,5-(1′,3-di-2-thienyl-5′,7-bis(2-

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ethylhexyl)benzo[1′,2′-c:4′,5′-c′]dithiophene-4,8-dione))] (PBDB-T) and 3,9-bis(6-methyl-2-methylene-(3-(1,1-dicyano methylene)-indanone))-5,5,11,11-tetrakis(4-hexylphenyl)-dith ieno[2,3-d:2′,3-d′]-s-indaceno[1,2-b:5,6-b′]dithiophene (IT-M). The PBDB -T:IT-M solar cells were stored in an argon-filled glove box under dark and indoor light conditions for 40 weeks. The current density‒voltage (J‒V) curves of solar cells were measured to confirm the influence of different storage environment on the long-term shelf lifetime of solar cells.

2. Experimental Section

2.1 Materials and solutions

Zinc oxide (ZnO) precursor solutions were prepared by dissolving 100 mg zinc acetate dihydrate in 1 mL 2-methoxyethanol and 0.028 mL ethanolamine (stabilizer). The solution was first stirred at 60 °C for 3 h and then again at room temperature for 12 h to proceed to the sol‒gel process. All reagents were obtained from Sigma-Aldrich (USA). The blend solutions of PBDB-T (weight‒average molecular weight = 18 kDa; polydispersity index = 2.5) and IT-M (molecular formula = 1.455 kDa) purchased from Solar Materials Inc. (China) were prepared using chlorobenzene (CB), including 1,8-diiodooctane (DIO, 1 vol%) as an additive, at a solid concentration of 20 mg/mL (PBDB-T: IT-M = 1:1 by weight). All solutions were stirred at room temperature for 24 h to prepare uniform thin films by spin-coating.

### 2.2 Device fabrication and measurement

To fabricate inverted-type PBDB-T:IT-M solar cells, pre-patterned ITO-coated glass substrates were cleaned with acetone and isopropyl alcohol using an ultrasonic cleaner. The cleaned ITO-coated glass substrates were treated inside a UV-ozone cleaner (28 mW/cm^2) for 20 min to remove some organic residues. The ZnO precursor solutions were spin-coated on the cleaned ITO-glass substrates and then underwent thermal annealing at 200 °C for 1 h in ambient condition. The PBDB-T:IT-M solutions were spin-coated on the ZnO layers, which were progressed in a nitrogen-filled glove box system. Thereafter, the coated samples were placed in an argon-filled vacuum chamber for electrode deposition. The molybdenum trioxide (MoO$_3$, 10 nm) and silver (Ag, 80 nm) electrodes were thermally evaporated on the PBDB-T:IT-M layers in the chamber with a base pressure of ca. 1.0×10^{-6} torr.
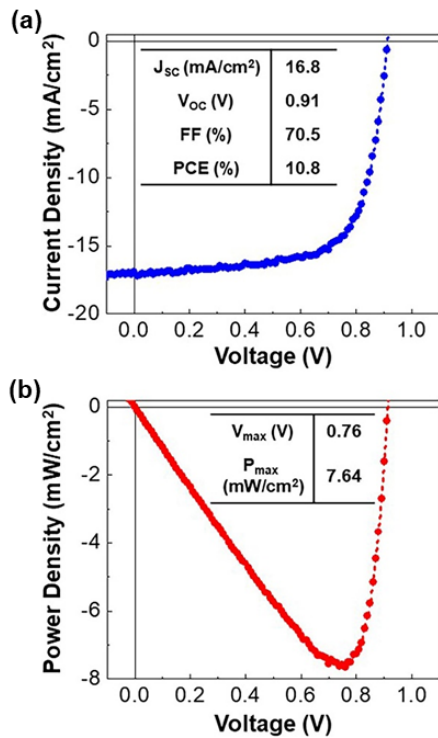
The fabricated solar cell samples were stored in an argon-filled glove box under dark and indoor light conditions to measure their long-term storage stability. The J‒V curves of solar cells were measured using a solar cell measurement system equipped with a solar simulator (92250A-1000, Newport Oriel) and electrometer (Keithley 2400). The fixed solar light intensity, 100 mW/cm^2 (1 sun, air mass 1.5 G), was confirmed using a calibrated standard cell (BS-520, Bunkoukeiki Co. Ltd). The surface of the films and solar cells was examined using an optical microscope (SV-55, SOMETECH). The optical absorption spectra of coated films were measured using a UV‒visible spectrometer (Lambda 750, PerkinElmer).

3. Results and Discussion

As shown in Fig. 1, the present PBDB-T:IT-M solar cells were fabricated with an inverted-type geometry. Note that four sub-cells were formed on one substrate by the deposition of Ag
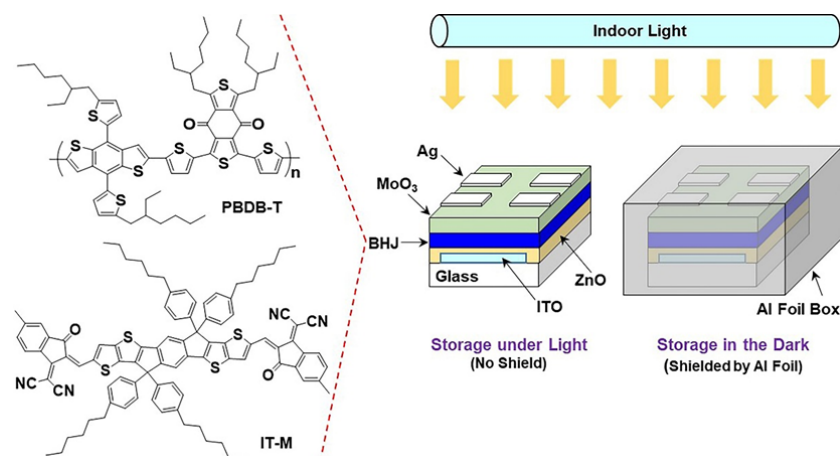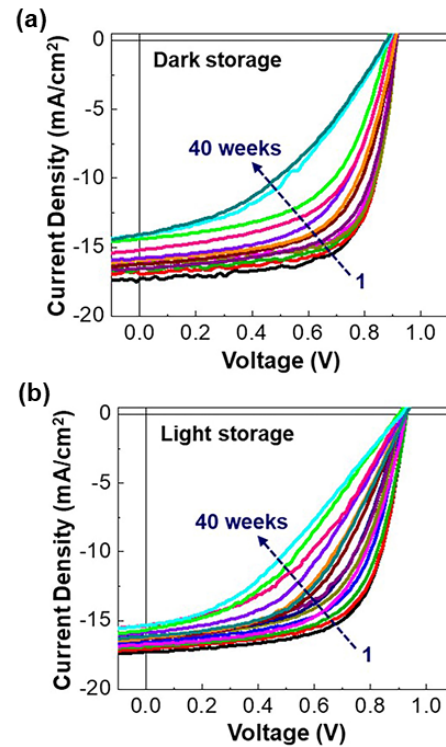

Fig. 1. Chemical structure (left) of the materials used for the BHJ layers in the inverted-type PBDB-T:IT-M solar cells and illustration (right) for the storage experiment of the PBDB-T:IT-M solar cells

(a)

(b)

Fig. 2. Light J−V curve (a) and power density−voltage curve (b) of the fresh PBDB-T:IT-M solar cells

(a)

(b)

Fig. 3. Light J−V curves of PBDB-T:IT-M solar cells stored under dark (a) and indoor light (b) conditions for 40 weeks

electrodes. To measure the long-term stability of the fabricated PBDB-T:IT-M solar cells stored under dark and indoor light conditions, the solar cells were stored in an argon-filled glove box, with one set covered with aluminum foil to block the indoor light and the other set exposed to the indoor light. The PBDB-T:IT-M solar cells were measured every week for 10 months (40 weeks).

As shown in the light J−V curve of fresh solar cell (0 week) (see Fig. 2 (a)), the best PCE reached 10.8%. The short-circuit current density (J_{SC}) and open-circuit current voltage (V_{OC}) values of the present PBDB-T:IT-M solar cells were similar to those reported in our previous reports[38]. In addition, the fill factor (FF) of 70.5% supported that the physical leakages could be minimized in the device fabrication process. As shown in the power density−voltage (P−V) curve (see Fig. 2 (b)), the maximum power density (P_{max}) and voltage (V_{max}) reached 7.64 mW/cm^2 and 0.76 V, respectively. The light J−V curves of solar cells stored under the dark condition are shown in Fig. 3 (a). The shape of J-V curves became gradually poor as the number of week increased, implying that the performance of solar cells was reduced with the time. The initial PCE of 10.83% decreased to 4.93% after 40 weeks. However, it is worthy to note that the PCE was kept higher than 10% for 13 weeks without an obvious change in the V_{OC} value even though the J_{SC} value gradually

decreased from 17.37 to 15.9 mA/cm^2. At 39 weeks, a noticeable change in the J_{SC} value significantly impacted the FF, eventually leading to a decrease in PCE from 10.83% (1 week) to 4.93% (40 weeks), although the V_{OC} value slightly changed from 0.91 to 0.87 V. The light J−V curves of the solar cells stored under indoor light conditions in Fig. 3 (b) exhibited a similar J_{SC} and V_{OC} reduction trend compared with Fig. 3 (a). However, the V_{OC} value was maintained over 0.9 V for 40 weeks without a significant change, whereas the J_{SC} value gradually decreased from 17.32 mA/cm^2 (1 week) to 15.29 mA/cm^2 (40 weeks). As a consequence, the PCE of solar cells was changed from 10.54% at 1 week to 4.84% after 40 weeks.

The P−V curves of the solar cells based on the storage condition are demonstrated in Fig. 4. As shown in Fig. 4 (a), the P−V curves of the solar cells stored under dark conditions showed a change in the maximum power density and voltage when the storage time was increased up to 40 weeks. The maximum power density slightly decreased from 7.42 mW/cm^2 (1 week) to 6.95 mW/cm^2 (15 weeks) without a conspicuous change in the maximum voltage. After 18 weeks (6.32 mW/cm^2), the maximum power density drastically decreased at 36 weeks (3.93 mW/cm^2), whereas the maximum voltage changed from 0.73 to 0.63 V. A significant reduction of 2.68 mW/cm^2 was observed at 39 weeks, which eventually decreased to 1.98
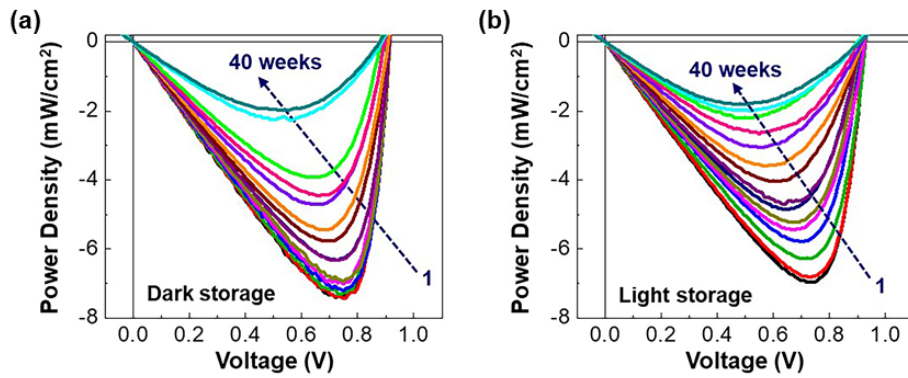
Fig. 4. Power density–voltage curves of PBDB-T:IT-M solar cells stored under dark (a) and indoor light (b) conditions for 40 weeks
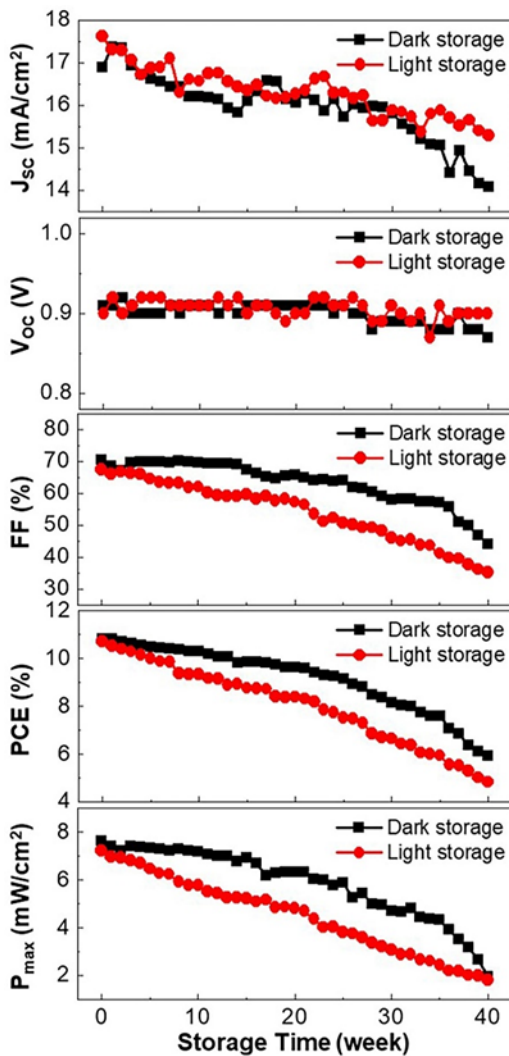

Fig. 5. J_{SC}, V_{OC}, FF, PCE, and P_{max} as a function of the storage time (week) for the PBDB-T:IT-M solar cells

mW/cm^2 after 40 weeks (V_{max} = 0.5 V). The maximum power density of solar cells stored under indoor light conditions gradually decreased from 6.97 mW/cm^2 at 1 week to 1.81 mW/cm^2 after 40 weeks, as shown in the P–V curves of Fig. 4

(b), whereas the maximum voltage changed from 0.73 V at 1 week to 0.49 V after 40 weeks. In order to further understand the influence of the long-term storage of solar cells under dark and indoor light conditions, the parameters of solar cells are plotted in Fig. 5 and summarized in Table 1. The J_{SC} value of the solar cells stored under dark conditions for 40 weeks decreased from 16.90 mA/cm^2 (0 week) to 14.17 mA/cm^2 (40 weeks), with a reduction rate of 16.2%, whereas the V_{OC} value changed from 0.91 to 0.87 V, with a reduction rate of 4.4%. For solar cells stored under indoor light conditions, the J_{SC} value changed from 17.62 mA/cm^2 at 0 week to 15.29 mA/cm^2 after 40 weeks, with a reduction rate of 13.2%, whereas the V_{OC} value slightly changed from 0.90 to 0.89 V at 40 weeks. As shown in Table 1, although the V_{OC} value did not have a significant impact on the change in PCE for 40 weeks, a noticeable reduction in J_{SC} and FF, including a change in R_S and R_{SH}, reduced PCE from 10.84% to 5.91% under dark conditions and 10.71% to 4.83% under indoor light conditions. However, the maximum power density was confirmed a similar value after 40 weeks, regardless of the storage condition.

The surface characteristics of the BHJ (PBDB-T:IT-M) layers in the solar cells stored under dark and indoor light conditions for 40 weeks were measured using an optical microscope (Fig. 6 (a)). Before being stored under dark and indoor light conditions, the fresh film (0 week) had a clean surface. However, after 40 weeks, black spots appeared. These black spots can be attributable to the degradation of molecules in the BHJ layers, which affected the FF and J_{SC} reduction and eventually resulted in the lowered PCE after 40 weeks. The solar cells stored under indoor light conditions exhibited lower efficiency than those stored under dark conditions after 40 weeks because of more than twice as many black spots generated on the PBDB-T:IT-M layers. The generation of black spots can be attributable to the degradation of IT-M molecules

Table 1. Summary of solar cell parameters according to the storage condition (up to 40 weeks) for the PBDB-T:IT-M solar cells

Parameters	Storage Time (week)											
	0		8		16		24		32		40	
	Dark	Light	Dark	Light	Dark	Light	Dark	Light	Dark	Light	Dark	Light
V_{OC} (V)	0.91	0.90	0.90	0.91	0.91	0.91	0.90	0.91	0.89	0.89	0.87	0.90
J_{SC} (mA/cm^2)	16.90	17.62	16.44	16.30	16.33	16.48	16.14	16.29	15.43	15.72	14.17	15.29
FF (%)	70.51	67.51	70.22	61.86	66.34	58.32	63.91	52.25	58.41	45.54	44.19	35.17
PCE (%)	10.84	10.71	10.39	9.35	9.86	8.74	9.28	7.74	8.01	6.37	5.92	4.83
R_S (k$\Omega\cdot$cm^2)	0.05	0.06	0.05	0.08	0.07	0.12	0.08	0.20	0.12	0.31	0.51	0.38
R_{SH} (k$\Omega\cdot$cm^2)	1.93	5.88	1.69	9.58	4.35	5.60	1.30	4.18	9.83	2.42	2.40	0.90
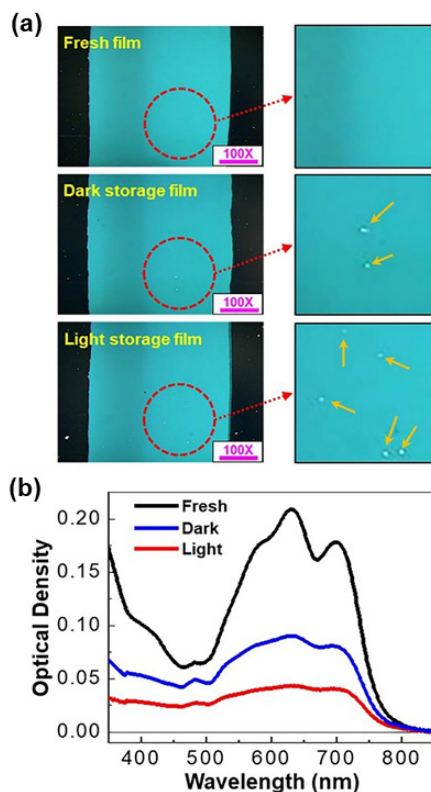
Fig. 6. (a) Optical microscope images and (b) UV–visible absorption spectra for fresh BHJ film (0 week) and the BHJ films (after 40 weeks) stored under dark and indoor light conditions

caused by the photo-induced cleavage of double bonds under continuous illumination of room light. Here it is noted that very small amount of oxygen molecules, whether they come from the argon flow in the glovebox and/or slowly extracted from the oxide materials (ZnO and MoO$_3$) in the devices, might affect the photo-induced degradation of IT-M[38,39]. The cause of the reduced efficiency of solar cells stored for 40 weeks was also confirmed in the optical absorption properties in Fig. 6 (b). Before storage, the absorption peaks of the PBDB-T:IT-M films (layers) were distinctly separated at 660 and 700 nm with a higher optical density. However, after 40 weeks, the PBDB-T:

IT-M films exhibited a lowered optical density in the absorption peaks with a lumped shape at each peak. The optical density of the films (active layers) stored under indoor light conditions decreased more than that under dark conditions, which reduced the efficiency for solar cells stored under dark and indoor light conditions for 40 weeks. The lower optical absorption property of the films (active layers) stored under indoor light conditions seemed to have been influenced by the UV light under indoor light condition, as reported in the previous work[38,39].

4. Conclusions

The long-term storage stability of the inverted-type PBDB-T: IT-M solar cells was measured for 40 weeks, depending on the environment in which they were stored under dark and indoor light conditions inside an argon-filled glove box. With respect to the performance of the solar cells, the J_{SC} and FF values gradually decreased when the storage time increased up to 40 weeks, and the V_{OC} value did not relatively have a significant change. The noticeable change in the FF and J_{SC} values eventually reduced the efficiency of solar cells after 40 weeks from 10.84% (0 week) to 5.92% (40 weeks) under dark conditions and from 10.7% (0 week) to 4.83% (40 weeks) under indoor light conditions. The reduction in the efficiency of solar cells stored under dark and indoor light conditions for 40 weeks was caused by the degradation of BHJ layers, as evidenced by the reduced optical density and dark spot growth. These results can draw a conclusion that the PBDB-T:IT-M solar cells stored under dark conditions for 40 weeks exhibited a higher efficiency than those stored under indoor light conditions.

Acknowledgments

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