

A study for degradation of flexible organic photovoltaic modules via damp-heat test: By accessing individual layers of the module

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ARTICLE INFO

Article history:

Received 6 May 2015

Received in revised form

26 July 2015

Accepted 31 August 2015

Keywords:

Organic photovoltaic module

Degradation

Damp heat test

Ag-migration

Lifetime pattern.

ABSTRACT

The stability of slot-die coated flexible organic photovoltaic (OPV) modules with inverted structures of indium tin oxide (ITO)/ZnO/photoactive layer/poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS)/Ag was analyzed under damp heat conditions. The work primarily focused on the understanding of the degradation mechanism of organic photovoltaic modules in very severe operating conditions (85 °C/85% relative humidity [rh]). The electrical change of the OPV module was measured as a function of the aging time under damp heat. A rapid drop in the efficiency of the module was observed, mainly caused by the decline of the open-circuit voltage (V_{oc}) and fill factor (FF). We supposed that the degradation of the modules mainly progressed by the oxygen and moisture penetrating the terminals or at the edges of the barrier films. The physical modification of the layers composing the device by degradation was observed; hence, morphological and chemical analyses were conducted. The analyses revealed that Ag atoms migrated to the interlayers of the cell, resulting in the increase of shunt paths; this was the main reason underlying the reduction of V_{oc} and FF of the OPV module under damp heat. The ZnO layer was also etched by the acidic molecules from the diffused PEDOT:PSS polymer. In addition, the performance parameters of several OPV modules were evaluated simultaneously under damp heat conditions for 1000 h. The same lifetime patterns were observed although the initial efficiencies of the modules were diverse. The interpretation of the lifetime patterns and a suggestion for an objective comparison between the modules showing different initial efficiencies were also addressed.

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1. Introduction

Numerous studies on organic photovoltaics (OPV) have been performed and progressive improvements have been achieved in the photo conversion efficiency using bulk hetero-junctions of poly(3-hexylthiophene) (P3HT) and 1-(3-methoxycarbonyl)-propyl-1-phenyl-(6,6)C61 (PCBM) blends and new donor materials [1–3]. Many merits of OPVs, such as light weight, low-cost production, and flexibility, raise the competitiveness of the technology in the market. Printing processes have been considered excellent fabrication methods that enable the large-area production of flexible OPVs without vacuum processing systems. Hence, various printing technologies have been widely

investigated, including slot-die coating, doctor blading, and screen printing [4,5].

However, several limits remain that should be overcome, compared to other types of solar cells. For commercialization, the OPV module is urgently requires stability improvement, since the organic materials are relatively sensitive to outdoor conditions. The degradation typically progresses via the reaction of the layers composing the device with oxygen and water. Related studies have confirmed the mechanisms underlying chemical and physical degradation: penetration of oxygen and water in the device degrades each layer of the cell or the interfaces between layers [6–8]. As a result, the device experiences a serious decline in the power conversion efficiency, accompanied with a change in appearance. Therefore, the protection of the device from atmospheric elements has been a main concern. Remarkable achievements in the stability of OPV modules have been accomplished with the application of various encapsulation materials [9–11].

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Rigid and flexible barriers such as glass or poly(ethylene terephthalate) films were used; the devices showed outstanding stabilities for long periods. The atmospheric elements can diffuse through not only the barrier materials but also the edges of the device [12,13]. Therefore, additional sealing around the edges further slowed the progress of degradation [14]. Rösch and coworkers mapped the degradation appearance by imaging methods. Several causes of performance degradation were identified, including electrode oxidation, blocking layer formation, electro-migration of Ag, and water released from highly conductive poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) [15]. The terminals connecting to the external circuit can also provide diffusion paths for oxygen and water. The presence of water near the terminals was observed; the water was likely to spread into the device [9].

Some accelerating tests for OPV modules have been employed for the fast evaluation of the lifetime of the cell. The properties of the electrode applied to the device can vary, affecting the lifetime. The application of a Ca/Ag cathode to the solar cell enhanced the lifetime under accelerated conditions (100 mW/cm² illumination, 60 °C) corresponding to a lifetime of 1000 h under real conditions [16]. Replacing the bottom electrode with other metal complexes showed remarkable increases in stability [17]. The experiments were carried out under different ageing conditions, including damp heat, thermal stress, and light soaking. The module performance losses were less than 10% after 1800 h under damp heat and 20% for 12,000 h with light soaking. The main factor that degraded the performance of the device under damp heat conditions was the corrosion of the electrode, which resulted in the increase of the series resistance of the cell.

In this work, we conducted an accelerating test of OPV modules encapsulated with flexible barrier films under damp heat conditions. The reasons affecting the degradation of the device were elucidated through electrical characterization and microscopic analysis. To specifically understand the degradation mechanism, morphological and chemical analyses were conducted for each layer of the cell. The degraded part of the surface of the layers was observed and locally measured to obtain compositional information. Moreover, simultaneous damp heat testing for several OPV modules was also conducted in order to compare the degradation patterns according to the various initial efficiencies of the modules.

2. Experimental

The organic photovoltaic modules were fabricated as striped cells with the inverted structure of indium tin oxide (ITO)/ZnO/photoactive layer/PEDOT:PSS/Ag. The OPV cells were slot-die coated on flexible ITO (200 nm & 30 ohm/sq)/ PET, with the exception of the screen printing of the Ag paste electrodes (10 μm), according to the fabrication process described by another group [18]. The photoactive material (200 nm) and PEDOT:PSS (4 μm) were purchased from Merck and Agfa (Orgacon EL-P 5010), respectively. The total functional area of the nine solar cells aligned in series was approximately 42 cm². The 200 nm thick SiO_x barrier layer deposited PET film by plasma-enhanced chemical vapor deposition (PECVD) was press-laminated to the front and back sides of the cells with optically clear adhesive (OCA) (50 μm). The permeation rate of the barrier deposited PET film for water is ~2.61 mg/m²-day (measured according to ASTM F372-78). The water vapor permeation rate was measured in both the transient and steady-state regimes of PET with a barrier layer. There is a buffer of approximately 1 cm width around the four sides of the square between the active cells and the edges of the covering barrier film on the module. This buffer could also affect the penetration rate for water and oxygen through the edges. Accordingly, we applied the same length of buffer area over all testing modules. The terminals acting as external contacts were in the shape of metal snap fasteners.

Several OPV modules were stored in a dark, high-temperature, and humid atmospheric chamber (85 °C/85% rh). Visual inspections for changes in appearance and electrical analyses of the performances of the modules were conducted every ~50 h. IV measurements for each OPV module were performed using a Keithley 2400 source meter and WACOM (WXS-90S-5) solar simulator under an illumination level of 1000 W/m² intensity with an AM1.5G optical filter. To calibrate the light radiation level, a reference device of a silicon detecting cell was used prior to the IV measurements. To further confirm the physical and chemical modification of the OPV cells, X-ray photoelectron spectroscopy (XPS), scanning electron microscopy (SEM), and energy-dispersive X-ray analysis (EDX) were performed after the damp-heat test. Processes for accessing each layer of the device are described in Fig. S1 (see the Supplementary information). The sheet resistance of each layer in the device was measured using a four-point probe device.

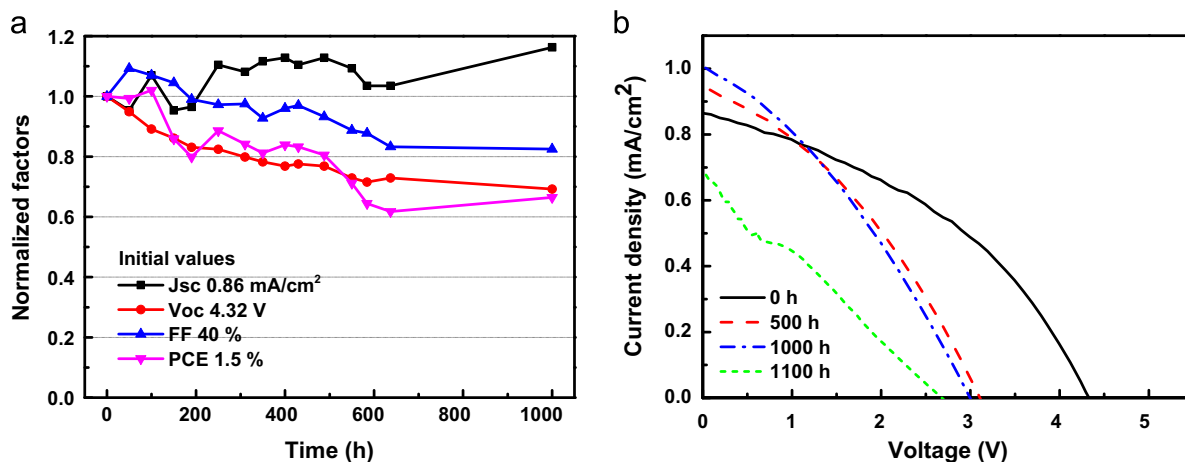


Fig. 1. (a) Normalized performance factors of a flexible OPV module as a function of the aging time under damp heat conditions (85 °C/85% rh) and the initial values of the device. (b) Time-evolution J - V (Current density-voltage) curves of the same module.

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