



A stability study of roll-to-roll processed organic photovoltaic modules containing a polymeric electron-selective layer



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ARTICLE INFO

Article history:

Received 1 February 2016

Received in revised form

7 March 2016

Accepted 24 March 2016

Available online 12 April 2016

Keywords:

PEIE

Organic photovoltaics

Encapsulation

Barrier films

Stability

ABSTRACT

The stability of roll-to-roll processed organic photovoltaic modules having an inverted structure and incorporating polyethylenimine–ethoxylate (PEIE) as the electron-selective layer was investigated. Large-area modules were fabricated on ITO-coated PET substrates using roll-to-roll coating and printing methods. Modules were encapsulated with commercially-available ultra-high gas/vapor barrier films by employing new encapsulation protocols developed during this work. The operational lifetime of modules on storage in an environmental chamber at ambient temperature and relative humidity of 35 °C and 50%, respectively, and under continuous simulated AM1.5G illumination was found to increase by more than three orders of magnitude upon encapsulation. The chemical stability of the PEIE films under these storage conditions was also studied. Fracture tests were conducted on modules exposed to the same storage conditions to investigate the effects on inter- and intra-layer adhesion and cohesion. An increase in adhesive strength was found for the exposed devices indicating an absence of any substantial mechanical degradation of the deposited layers for the exposure conditions used during this work. The results described here demonstrate the potential utility of commercially-available PEIE as a convenient and effective organic electron-selective layer for the fabrication of durable roll-to-roll processed organic solar cell devices.

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1. Introduction

Organic photovoltaic (OPV) technology has been the focus of considerable research and development effort because of the unique potential benefits offered for a wide variety of commercial applications. Significant progress has been made in the improvement of the power-conversion efficiency (PCE) of polymer bulk-heterojunction (BHJ) solar cells over the past two decades, with PCE values in excess of 11% having been reported recently for small laboratory-scale tandemly structured devices [1]. One key advantage of OPV technology is that all of the functional layers can be deposited on flexible plastic substrates using environmentally-benign, scalable, roll-to-roll (R2R) coating and printing technologies [2–4]. Furthermore, the simplicity of the fabrication process is expected to enable very low-cost manufacturing of flexible and lightweight OPV modules for applications such as building-integrated PV systems [5]. Recent progress in the development of large-area flexible solar cell modules has resulted

in the demonstration of large-scale, continuous manufacturing of OPV modules and also the potential of OPV technology to revolutionize the use of solar energy in the very near future [2,6,7].

However, apart from requiring adequate PCE, sufficiently long operational lifetime is also essential for real-world applications. Instability in the performance of OPV modules upon exposure to environmental factors such as oxygen, elevated temperature and relative humidity, and UV irradiation remains a major limiting factor for the successful commercialization of flexible OPV technology [8]. Thus, a key research challenge is to overcome this poor stability to achieve the necessary operational lifetime for intended end uses. Understanding the degradation mechanisms of flexible OPV systems, and extended lifetime studies to monitor module performance under real-world conditions, are crucial for realizing the successful commercial application of flexible OPV technology.

Recent studies by our group and others have shown that integration of suitable barrier encapsulation can significantly improve the environmental stability of OPV modules fabricated using ZnO as the electron-selective layer [9,10]. The commercially-available, highly-branched polyamine, polyethylenimine–ethoxylate (PEIE), has recently been applied in conjunction with ZnO to OPV devices

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to increase the efficiency of inverted cells by reducing the work function of the cathode, thereby reducing the trap-assisted recombination [11]. It has also been shown that PEIE alone can be used as an alternative electron-selective layer to prepare efficient small-scale organic photovoltaic devices [12]. Furthermore, PEIE has also been applied in highly efficient tandem OPV devices as an excellent recombination layer [13,14]. Compared with the electron-transporting metal oxides such as ZnO or TiO₂ that are commonly used in the fabrication of flexible OPV solar cells, PEIE provides the prospect of a more convenient, cost-effective, organic-based alternative that is compatible with the fabrication of large-area OPV modules via low-temperature R2R solution deposition on flexible substrates. However, to date, the preparation and systematic study of the stability of large-area R2R processible OPV modules using PEIE as the electron-selective material has not been reported.

In the present work, we present the results from a preliminary investigation of the stability of large-area (100 cm²) R2R processed OPV modules using PEIE as an organic electron-selective layer. The effect of exposure to continuous 1-sun irradiation and moderate ambient temperature and relative humidity, or vigorous bending, on the performance of encapsulated and un-encapsulated modules was evaluated, with PV performance results and mechanical analysis used to elicit information relating to the failure mechanism of the R2R processible modules.

2. Experimental

2.1. Device fabrication and encapsulation

R2R processed OPV modules comprising five series-connected strip cells in an “inverted” configuration were fabricated on an ITO-coated PET film (15 Ω/□, LRS15, Solutia) to give a device consisting of PET/ITO/PEIE/P3HT:PCBM/PEDOT:PSS/Ag (Fig. 1(A)). 13 mm wide ITO pattern with 2 mm wide gaps were used, and for the electrical contacts, extended first ITO stripe and an extra ITO stripe on the other end was used [15,16]. An aqueous solution of PEIE (Mw 70,000 g/mol, 35–40% w/w, Aldrich) was diluted with 2-methoxyethanol to a concentration of 0.15% w/w. This solution was coated onto ITO-coated PET film by the reverse-gravure coating method using a Mino-Labo™ coater (Yasui Seiki Co. Ltd) to form a PEIE film having an estimated thickness of around 10 nm. All the layers (PEIE, P3HT:PCBM and PEDOT:PSS) deposited on the reverse-gravure coater were coated on ITO-PET substrates using a 200 R roll with a 5 × 13 mm² stripe pattern having 2 mm gaps. Coating was carried out at 0.36 m/min speed and 8 rpm of roll speed. The PEIE layer was dried by hot air at 110 °C for 20 s and annealed on a curved hot plate at 140 °C for 20 s. The P3HT (SP001, Merck):PCBM (Tec. Grade, Solenne) (poly(3-hexylthiophene):phenyl-C61-butyric acid methyl ester) (~200 nm thick) and PEDOT:PSS (S315, Agfa) (poly(3,4-ethylene dioxythiophene):poly(styrene sulfonate)) layers (~1 μm thick) were coated at 0.36 m/min web speed and 10 rpm of roll speed. During the active layer deposition, film was dried only using hot air at 70 °C for ~20 s, and PEDOT:PSS deposition was carried out with a drying step using hot air at 110 °C and on a curved hot plate at 130 °C for ~20 s. A screen-printed grid using a commercially-available Ag ink (DuPont, PV416) was used to enhance charge collection of the module and to interconnect the 5 cells in series. Screen printing was carried out manually as well as on an Orthotec-2 R2R screen printing system. Each deposited layer had 0.2–0.5 mm offset from the under-laying layer so that series-connection could be made. The screen-printed Ag grid was dried at 130 °C for 30 s on a hot plate. The active area of each strip cell is 10.5 cm² (width:1.1 cm and length: 9.5 cm) resulting an active module area of ~52.5 cm².

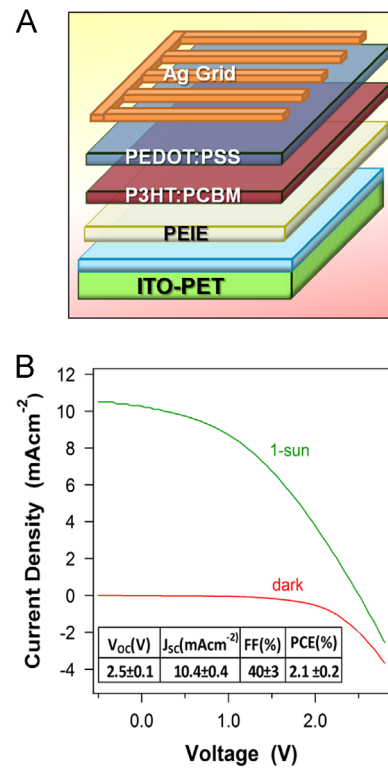


Fig. 1. (A) Schematic representation of the structure of the PEIE-based OPV modules. (B) *J*–*V* characteristics measured for a PEIE-based OPV module under simulated solar radiation (100 mW cm², AM1.5G) and in the dark. Average *J*–*V* parameters for 50 modules are shown in the table.

The R2R processed structures used for mechanical fracture analysis, PET/ITO/PEIE/P3HT:PCBM/PEDOT:PSS, PET/ITO/PEIE/P3HT:PCBM, and PET/ITO/PEIE, were prepared in the same manner as the complete modules. PEIE films on glass substrates (glass/PEIE) were prepared by diluting the aqueous PEIE solution with 2-methoxyethanol to 4% w/w followed by spin coating to form PEIE films having a thickness of approximately 100 nm.

Prior to encapsulation, modules were pre-conditioned by storage under vacuum at room temperature for at least 12 h, followed by storage in a N₂-filled glovebox until the encapsulation process was carried out. All the encapsulant materials were pre-conditioned by storage under vacuum at 100 °C for 12 h [17]. Modules were encapsulated using architectures that will be referred to in this work as ‘partial’ and ‘complete’, as illustrated in Fig. 2, using commercially available ultra-high gas barrier films and barrier adhesives. For the simpler, ‘partial’ encapsulation architecture, 3M™ Ultra Barrier Solar Film layers were laminated onto the front and back sides of the module using a transfer adhesive layer (3M™ 467MP), with the barrier film layer on the back side being slightly narrower than the module in order to allow direct electrical contact to be made with the device electrodes (Fig. 2(A)). As illustrated in Fig. 2(B), ‘complete’ encapsulation consisted of 3M™ Ultra Barrier Solar Film encapsulant layers laminated onto the front and back faces of the module using ethylene-vinylacetate (EVA) sheets (~100 μm thick) as adhesive interlayers. HelioSeal® PVS 101 tape (ADCO) was applied around the perimeter (~1.3 cm wide) of the modules for additional moisture-barrier edge-sealing. Electrical contact to the module was made via 100 nm-thick Ag metal strips evaporated onto the inner face of the back barrier encapsulant film which were connected to the module electrodes using z-axis conducting adhesive (3M™ Electrically Conductive Adhesive Transfer Tape 9703). The evaporated metal strips were covered by the

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