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Influence of an electrode self-protective architecture on the stability of inverted polymer solar cells based on P3HT:PCBM with an active area of 2 cm^2

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ABSTRACT

In this paper, we study the performance and stability of solution-processed inverted organic solar cells based on photoactive blends composed by the conjugated regioregular poly-(3-hexylthiophene) (P3HT) and [6,6]-phenyl-C₆₁-butyric acid methyl ester (PCBM), using an active area of 2 cm^2 . These inverted organic solar cells are fabricated with a novel top electrode design in which the silver electrode is deposited over the whole substrate to completely cover the photoactive layer, allowing an effective protection of the entire device. Consequently, initial power conversion efficiencies of 3.2% are maintained at 90% after 15 h under standard illumination conditions in ambient atmosphere. Light beam induced photo-voltage (LBIV) maps have been recorded to monitor the uniformity of the photo-response on the whole active area, and revealed the effectiveness of our design to prevent lateral moisture and oxygen diffusion. The dependency of short-circuit current density on incident light intensity, combined with LBIV data, indicates that the degradation of performance arises from dark spots which reduce the active layer area, rather than from an intrinsic aging of the active layer. Taking into account these observations, we rationalize the time evolution of device efficiency upon degradation for both non-encapsulated and encapsulated devices.

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1. Introduction

In the last decade, an intensive research has been carried out in the community to understand the degradation mechanisms [1-4] and improve the stability of organic or polymer solar cells [5,6]. Stability is an essential issue in polymer solar cells for achieving commercially applicable devices [7]. Inverted organic solar cells (IOSCs) show an attractive architecture associated to long lifetime and an ability to reduce the degradation of performance [8], owing to the relative environmental stability of the metal oxide electron transport layer (e.g ZnO,TiO₂) and relatively high work function Ag hole-collecting contact. In general, degradation of organic solar cells (OSCs) is mostly caused by diffusion of water and moisture in the device active layer through pinholes or grain boundaries introduced at the outer electrode [2,9–11]. In addition, the deposition of metal often leads to atom diffusion in the active layer, which can induce chemical reaction with the polymer and alter its semiconducting properties [1,12]. The degradation of

http://dx.doi.org/10.1016/j.synthmet.2015.12.021 0379-6779/© 2015 Elsevier B.V. All rights reserved. organic devices is generally determined by a two stage decrease of performance consisting of a fast initial drop, followed by a linear degradation. Such behavior was reported in a large number of articles [13–16]. Several strategies can be employed to improve the device stability, such as inserting buffer layers between the active layer and metallic contacts. The most effective approach is to adopt encapsulation techniques that prevent or limit the exposure of organic electronic devices to water and oxygen [17]. The best encapsulation process was reported in 2013, where the power conversion efficiency (PCE) maintained up to 94% of its initial value after 6145 h [18]. This encapsulation method remains however rather complicated since it uses atomic layer deposition (ALD) technique.

Beyond encapsulation techniques, little work has been reported on the design of devices to improve their stability. Particularly the sandwiched structure of OSCs presents an intrinsic weakness associated with the vertical and lateral diffusion of oxygen and water. Due to the imprecision inherent to roll-to-roll deposition methods, as well as to prevent electric leakages, the top electrode shows generally a smaller surface than the active layer and than the bottom transparent electrode. A significant portion of the





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Fig. 1. (a) Inverted organic solar cell architecture and (b) device layouts for the non-encapsulating and top electrode self-encapsulating architecture (TESEA) presented in this study.

active layer remains thus directly exposed to the outside atmosphere, leading to the ignition of diffusion mechanisms from the sides to the center. In this paper, we demonstrate efficient and stable inverted organic solar cells based on solution-processed P3HT:PCBM active layers and with a large active area of 2 cm². To achieve better device lifetime, we propose a new device layout based on an encapsulating metallic top electrode that was thought to reduce the direct exposure of the organic materials to ambient atmosphere (Fig. 1). The active layer is indeed entirely covered by the silver anode excepted at the edge comprising the ITO electrode contact.

Using such top electrode self-encapsulating architecture (TESEA), all devices show improved stability compared to cells fabricated with the conventional device configuration.

A comparison of the photovoltaic parameters of the cells under standard illumination in air (open circuit voltage V_{OC} , short-circuit current density J_{SC} , fill factor FF, and power conversion efficiency η) and their time-dependency, in addition to the consideration of the corresponding light beam induced voltage (LBIV) maps, allow us to discuss the reduced degradation of our TESEA devices compared to non-encapsulating references in terms of lateral diffusion of moisture or oxygen.

2. Experimental

For IOSC fabrication, $2.5 \times 2.5 \text{ cm}^2$ indium tin oxide (ITO)coated glass substrates purchased from Visiontek Systems Ltd. (Chester, UK) with sheet resistance of $7\Omega/\Box$ were patterned by wet chemical etching to define rectangular active area of 2 cm^2 . The substrate were then cleaned by successive sonications (10 min each) in deionized water, acetone, ethanol and isopropanol. After an additional UV-ozone treatment (10 min), gold and silver 60 nm thick stripes were deposited separately onto the substrates, to act as sub-electrodes in order to reduce the resistive losses due to ITO

[19]. They were respectively deposited by thermal evaporation and ion beam sputtering (IBS) [20-22] on the substrates through shadow masks. The silver stripe was deposited using an Ag target (99.99% purity) with the ITO substrate oriented at 60° with regard to the target axis, and placed at 6 cm away. Sputtering was carried out at a pressure of 4.10^{-4} Pa, using argon ions at 6 k eV, and with a current density of about 1 mA/cm^{-2} . In these conditions, the growth rate was in the order of 1.6 nm/min. Zinc oxide (ZnO) deposited by spin-coating from a nanoparticle suspension (ZnO 5F2) purchased from Genes'Ink (Rousset, France) and annealed at 130 °C for 10 min in air was used as electron injecting layer. The photoactive layer was formed from a blend solution of P3HT:PCBM (1:0.8 weight ratio) prepared from materials purchased from BASF (Germany) and American dye source Inc. (Quebec, Canada) respectively. P3HT and PCBM were dissolved in ortho-dichlobenzene (Sigma-Aldrich) and the solution was stirred inside the glove box at 50 °C overnight before being filtered (PTFE filters, $0.2 \,\mu m$) previously to film deposition. The active layer was spin-coated under nitrogen 780 rpm for 40 s and annealed at 50 °C for 15 min, leading to a 220 nm thick film. A 40 nm thick layer of PEDOT:PSS (RD CLEVIOS F010, Heraeus, Leverkusen, Germany) was spincoated on top of the P3HT:PCBM layer in air, after passing through a $0.45\,\mu m$ filter. Finally, a 100 nm thick Ag top electrode was deposited by thermal evaporation through a shadow mask at a pressure of 10^{-6} mbar to yield a device active area of 2 cm^2 (see Fig. 1). A final annealing step was performed at 110°C for 30 min under nitrogen.

Film thicknesses were measured using a Bruker Dektak XT profiling system [23]. The current density-voltage characteristics of the devices were recorded using a computer-controlled Keithley 2400 source-measure unit, in the dark and under simulated sunlight provided by an ATLAS Solar Constant 575PV solar simulator for measurements made under nitrogen, and by a NEWPORT (Class A) solar simulator for measurements made in



Fig. 2. Current density/voltage characteristics of fresh (a) and aged (b) devices recorded in the dark (dash lines) and under 100 mW cm⁻² (solid lines). The cell employing the encapsulating electrode corresponds to red traces, while the reference (non-encapsulating electrode) cell corresponds to black traces.

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