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Surface treatment patterning of organic photovoltaic films for low-cost modules

I. Etxebarria [a], J.G. Tait [b], R. Gehlhaar [b], R. Pacios [a,*], D. Cheyns [b]

[a] IK4-Ikerlan S. Coop., Goiru 9, Polo Innovación Garaia, ES-20500 Arrasate, Gipuzkoa, Spain
[b] Imec vzw, Kapeldreef 75, B-3001 Leuven, Belgium

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ABSTRACT

This work describes a patterning technique for the photoactive layer of organic photovoltaic modules. We demonstrate the fabrication of efficient poly[3-(hexyl)thiophene-2,5-diyl]:[6,6]-phenyl-C61-butyric acid methyl ester (P3HT:PCBM) based organic photovoltaic modules through a specific surface treatment, based on the deposition of a fluorinated self assembled monolayer (SAM) on top of the bottom electric contact. Direct self-patterning of the photoactive layer is achieved by the high contact angle between the SAM and the polymer solution, while a smooth topography is created by combining two solvents with different surface tensions and boiling points in the polymer:PCBM solution. The resolution of the patterning is approximately 400 μm for modules based on a conventional cell architecture and 120 μm for an inverted architecture. As a result, we show 25 cm^2 P3HT:PCBM based organic photovoltaic modules with 10 series-connected cells, fabricated via roll-to-roll compatible deposition and patterning techniques.

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1. Introduction

Organic photovoltaics (OPVs) based on solution processing of polymer:fullerene inks are attracting increasing attention in the industrial community. High efficiencies combined with the potential for low cost, easy integration into diverse applications, and large scale production presents the technology as a serious candidate to finally penetrate into the photovoltaic market. Therefore, organic photovoltaics are no longer a scientific curiosity, but an industrial objective with a few initial companies interested in commercialization and the first products already appearing on the market [1].

While record power conversion efficiencies for small area cells have attained 10.7% [2], larger area polymer devices suffer from efficiency-limiting losses, the most important loss being the low electrical conductivity of the transparent contact. In spite of these limitations, innovative applications based on roll-to-roll organic modules have been demonstrated [3–5]. Moreover, the business potential and market opportunities of polymer solar cells are very promising [6]. These opportunities rely on the demonstration of highly efficient modules fabricated via roll-to-roll compatible deposition and patterning techniques [7].

A general technique to overcome the limited conductivity of the transparent contact is by combining several smaller cells in a series connected chain. This ensures that the produced photocurrent of the overall module is limited, while the voltage increases linearly with the number of cells. The processing of these monolithically connected organic modules demands the patterning of each deposited layer in order to connect the cells in series (see Fig. 1). Spin coating [8] (not compatible with roll-to-roll), spray coating [9–11] and doctor blading [12] are all well known coating techniques from which continuous layers, i.e. without patterning, are obtained. In order to fabricate modules with any of these methods, additional patterning techniques like mechanical scribing [13] or laser scribing [14] are required. These methods, apart from the extra processing

* Corresponding author.
E-mail address: rpacios@ikerlan.es (R. Pacios).

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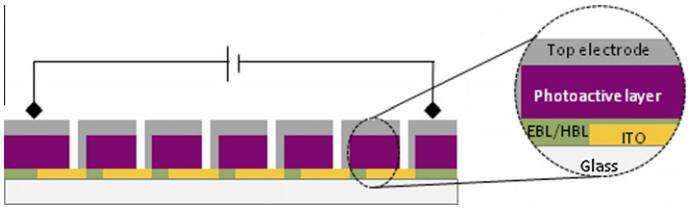

Fig. 1. Cross section of a series-connected module where the patterning required for each layer and the interconnections among the subcells can be seen.

step and increasing production costs, involve the risk of damaging the underlying layers and potentially the flexible foil that is commonly used as the holding substrate.

Instead, techniques like slot die coating [15,16] achieve simple patterned lines on the substrate. Moreover, with printing methods like screen printing [17], gravure printing [18] and flexographic printing [19] complex patterning can be obtained, making serial/parallel connections possible between single cells within the module. All of these techniques require specialized pieces of equipment, as well as the adaptation of deposition conditions for small area devices to those for large area devices. These conditions (primarily drying) define the bulk morphology of the polymer:fullerene film, and thus are critical to maximizing the power conversion efficiency of the final solar cell [20,21]. To avoid this tedious and time-consuming step we report a novel process that benefits from the advantages provided by spray coating and allows for direct patterning of the photoactive film.

In this paper we introduce a solution processable and roll-to-roll compatible patterning technology for the photoactive layer. Here, a fluorinated self-assembled monolayer is formed on top of the bottom contact prior to the deposition of the photoactive film. Thin lines of 1H, 1H, 2H, 2H-perfluorodecyltrichlorosilane (FDTS) are printed on the substrate with an inkjet printer. Due to the high contact angle between FDTS and the primary solvent (ortho-dichlorobenzene) used in the solution of the photoactive material, the liquid film is repelled from these lines, resulting in a fast and economic patterning of the photoactive layer.

2. Material and methods

Glass substrates (5.5×5.5 cm^2) with 10 individual ITO stripes (5 mm wide each with an area of 2.5 cm^2) and with a nominal sheet resistance of 20 Ω/square (SNP Taiwan Co.) were cleaned in subsequent ultrasonic baths of detergent, deionized water, acetone and isopropanol. Finally, the substrates were exposed to 15 min of UV ozone treatment in a UVOCS T10X10/OES Ultraviolet Ozone Cleaning System.

Afterwards, depending on the design structure, a hole injecting layer (for conventional devices) or electron injecting layer (for inverted devices) was deposited. In the case of conventional devices, 10 nm of MoO_3 was deposited by thermal evaporation at 10^{-7} Torr and a deposition rate of 1.2 Å/s. For inverted cells, a thin film of around 20 nm of zinc acetate (Zn(ac)) was spin coated on the substrate, which was subsequently baked at 300 °C in order to transform the Zn(ac) into zinc oxide (ZnO) [22].

A 0.5 mM FDTS solution was prepared using hexadecane as the solvent. FDTS lines were inkjet printed (Dimatix DMP-2831) on specific places of the substrates with the plate at 40 °C. The excess solvent was removed by annealing the sample at 200 °C for 5–10 s.

The regioregular P3HT (Rieke Metals, Inc. #4002-EE): PCBM (Solenne bv.) solutions were prepared with a ratio of 1:1 by weight. Different ratio blends of ortho-dichlorobenzene (ODCB) (Sigma Aldrich) and 1,3,5-trimethylbenzene (mesitylene) (Sigma Aldrich) were used to dissolve P3HT and PCBM. Solutions were stirred at 80 °C for at least 8 h and filtered (PTFE 0.5 μm) before processing.

The photoactive layer was deposited by spray coating (Sono-Tek Exactacoat equipped with an AccuMist 120 kHz ultrasonic atomizing nozzle) with a flow rate of 2.5 mL s^{-1} and nitrogen as a directing gas shroud. The substrate temperature varied from 55 °C to 80 °C depending on the module structure, solvent blend concentration and the photoactive film profile observed (see Results and Discussions section). The nozzle followed a raster pattern of parallel lines over the substrate with a 7 mm pitch. Immediately following the spray coating deposition, the samples were covered with a Petri dish to create a solvent saturated environment to slow the drying and create a more favorable morphology for charge generation and transport. Films were subsequently annealed in a nitrogen atmosphere at 130 °C for 10 min.

Finally, 20 nm of Yb [23,24] and 150 nm of Ag were thermally evaporated for conventional devices, while 10 nm of MoO_3 and 150 nm of Ag were deposited for inverted designs.

Before characterizing the modules, individual cells were connected in series by partially removing the top metallic contact from adjacent stripes of ITO by mechanical scribing.

The module characterization was performed in a controlled nitrogen environment with a Keithley 2602A Source-Measure Unit and an Abet solar simulator under 100 mW cm^{-2} AM1.5G illumination (class A), for a beam area of 25 cm^2. The calibration of the light intensity was performed using a certified Fraunhofer silicon PV cell, equipped with a band pass filter (KG3). Film thicknesses and profiles were measured by a Dektak D150 surface profilometer.

3. Results and discussions

Firstly, contact angle measurements were performed in order to analyze the adhesion properties of the FDTS as a function of the substrate treatment. Bare glass, ZnO and

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