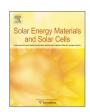
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Solar Energy Materials & Solar Cells

journal homepage: www.elsevier.com/locate/solmat



Fully roll-to-roll prepared organic solar cells in normal geometry with a sputter-coated aluminium top-electrode



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ARTICLE INFO

Article history: Received 28 October 2015 Received in revised form 7 December 2015 Accepted 12 January 2016

Keywords: Roll-to-roll processing Sputter coating Normal geometry Organic photovoltaics Printing

ABSTRACT

We demonstrate a pathway for fully roll-to-roll (R2R) prepared organic solar cells in a normal geometry with a R2R sputtered aluminium top electrode. Initial attempts utilizing a stack geometry without an electron transport layer (ETL) failed to obtain working devices. By applying aluminium zinc oxide (AZO) as an ETL, and optimizing the AZO thickness, working printed OPV devices with an efficiency of 0.58% were obtained. Further optimization of the donor:acceptor ratio in the active layer increased the efficiency to 0.90%. This work demonstrates that normal geometry organic solar cells using a metal top contact can be produced using large scale production techniques.

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1. Introduction

Solution-processed bulk heterojunction organic solar cells (OSCs) have received much interest from the research community during the last couple of decades due to their enormous potential as a low cost energy source [1,2]. The key-advantage of OSCs compared to conventional solar cells technologies is the capability of the materials to be solution processed by high throughput roll-to-roll (R2R) processing techniques; other advantages include low temperature processing and flexible substrates [3,4]. Even though the efficiency for small area (<0.1 cm²) devices has increased from approximately 2.5% in 2001 to 11.0% in 2014 [5], and that it is well-known that R2R techniques must be utilized for cost-effective fabrication of OSCs, only a few groups have turned their focus towards upscaling. Fabrication of large scale OSCs by R2R processing presents the unique challenge of generating uniform thin films with tailored morphologies and interfaces over stretches of hundreds of metres. By contrast, most laboratory scale OSCs are prepared by spin coating with an active area of < 10 mm² on a rigid glass substrate with indium tin oxide (ITO) as the transparent electrode [6]. Large scale production of

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OSCs uses a range of coating and printing techniques for ink deposition such as slot-die coating, flexographic printing and rotary screen printing [7,8]. Flexible substrates with a printed silver and coated poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS) electrode are commonly utilized for large scale OSC production to avoid the use of scarce materials such as indium [9,10]. The silver PEDOT:PSS electrode combination has been reported to obtain sheet resistance of $10.4 \,\Omega/\Box$ and is therefore comparable to ITO [9]. The most impressive OSC upscaling work to date was conducted by Krebs et al. [4] who produced a stretch of 21,000 defect free cells connected in series. This array of OSCs yielded over 10 kV under illumination with an overall performance of 1.5%.

The bulk of published work in the area of large scale OSCs prepared using R2R techniques has utilized inverted structure devices with a silver top electrode [9,11–14]. This structure represents a deviation in device geometry compared to laboratory scale devices, which typically have an evaporated electrode [6]; creating extra challenges on top of the already difficult process of upscaling [15]. Previous attempts to prepare normal geometry solar cells via R2R coating and printing techniques have all been with a non-scalable top electrode deposited by thermal evaporation [16–18]. Recently, Griffith et al. reported a procedure for sputter coating an aluminium top electrode onto OSCs [19]. In contrast to thermal evaporation, sputter coating is a widely used

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technique for the deposition of metal thin films on an industrial scale. There are three main advantages of a sputtered electrode over a printed silver electrode: 1) The sputtered back electrode is a reflective surface, allowing the preparation of thinner active layers whilst maintaining optical absorption, 2) OSC devices prepared in normal geometry exhibit enhanced $V_{\rm OC}$'s compared inverted geometry devices [20–22], 3) The harsh solvents and mechanical force present when flexographically printing or screen coating a top silver contact (which can lead to penetration of silver and solvent through previously coated layers to form areas with no photocurrent extraction [14,23]) are eliminated.

In this work we show the preparation and active layer optimization of flexible ITO-free organic solar cells by R2R coating and printing techniques using a R2R sputter-coated aluminium top electrode. This work demonstrates, for the first time, that normal geometry OSCs using a metal top contact can be produced using R2R techniques suited to large scale production.

2. Results and discussion

2.1. Device architecture optimization

Initial studies were conducted using cells with device geometry Structure A from Fig. 1 without an electron transport layer (ETL) as described in the literature [19] and with an active layer thickness of 500 nm. The active layer thickness was chosen to ensure that silver spikes found on the height profile in Fig. 1 were embedded in the active layer. The silver spikes had a height of approximately 250 nm, but larger spikes with a height of up to 350 nm were also present. Similar silver spikes have previously been observed on similar silver grid/PEDOT:PSS electrodes [24]. The prepared devices did, however, exhibit only limited solar cell characteristics as seen in Fig. 2; the obtained J-V curve has a slight S-shape with a FF of 23% and the J_{SC} is only 0.04 mA cm⁻². It has been suggested that the sputtering of metal onto soft organic layers can cause deep penetration of the metal due to the higher energy nature of sputtering as compared to thermal evaporation [25–27]. This phenomenon has been thought to cause shorts between the contacts in OSCs when sputtering has been employed for the top electrode. However, work by Griffith et al. has shown that, rather than causing deep penetration, the sputtering of aluminium on organic films forms an oxide layer near the active layer/sputtered electrode interface unless careful processing measures are

undertaken [19]. This oxide layer can have a detrimental effect on the performance of the OSC device.

In an attempt to overcome this issue, Structure B was utilized where an AZO layer was added as an ETL between the active layer and the sputtered aluminium electrode. AZO was chosen since it is a hard inorganic material (decreasing the likelihood of sputtering damaging the active layer) and no photoactivation is required, as is the case for example zinc oxide [28]. It was postulated that any residual oxygen present during the sputtering process should interact with the AZO layer rather than forming an oxide within the active layer. This hypothesis is supported by the J-V curves of devices containing an AZO layer as an ETL, which exhibit the desired J-shape (Fig. 2A) and a J_{SC} of 0.2 mA cm⁻², V_{OC} of 0.39 V

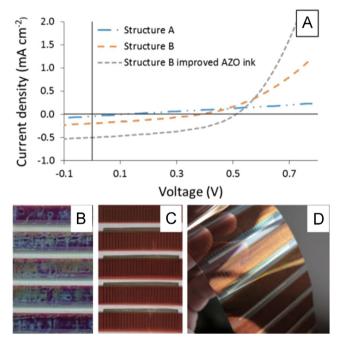


Fig. 2. (A) *J–V* characteristics of devices prepared with device Structure A, Structure B and Structure B with improved AZO ink, (B) Picture presentation of AZO layers prepared from a pure methanol AZO ink coated on top of active layer, (C) Picture of layer stack with optimized methanol:IPA AZO ink coated on top of active layer, and (D) Layer stack from picture B presented with light reflection. (For interpretation of the references to colour in this figure, the reader is referred to the web version of this article.)

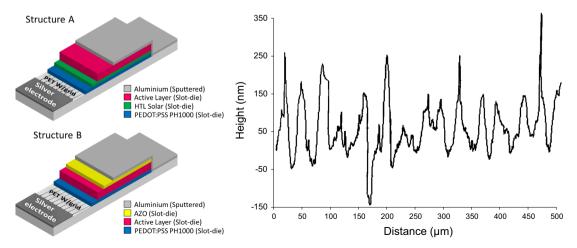


Fig. 1. Left: Two device structures were employed during the work presented in this paper along with deposition methods for the individual layers. Both device structures are in normal geometry but Structure A contains PEDOT:PSS as a hole transport (HTL) layer whereas Structure B contains aluminium doped zinc oxide (AZO) as an ETL layer. Right: Height profile of the silver PEDOT:PSS electrode along one of the silver fingers.

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