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Contents lists available at ScienceDirect

Solar Energy Materials & Solar Cells

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

Solution processing of back electrodes for organic solar cells with inverted architecture



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ARTICLE INFO

Article history:

Received 14 March 2014

Received in revised form

12 May 2014

Accepted 5 July 2014

Keywords:

Organic solar cell

Inkjet printing

PEDOT:PSS

Printed back electrode

Inverted cell architecture

Solution processing

ABSTRACT

Solution processing of the electrodes is a big challenge towards scaling up and R2R processing of organic solar cells. Inkjet printing is a non-contact printing method, it can be realized by solution processing at ambient condition and provides freedom of shape in the electrode pattern. The inkjet and screen printing of the back electrode in organic solar cells with inverted architecture is investigated. To this purpose, different types of PEDOT:PSS hole transport layers were combined with ambient processed inkjet printed or evaporated Ag electrodes. The thickness of the PEDOT:PSS layer is the main factor determining the performance of the devices. A too thin layer of EL-P5015 PEDOT:PSS allows penetration of the solvents from the Ag ink, which leads to degradation of the devices and a drop in performance. To overcome this effect, a 1200 nm thick layer of EL-P5015 is required. Alternatively, an aqueous based coating formulation of PEDOT:PSS (S305), can be combined with either inkjet printed or screen printed electrode at significantly reduced thickness down to 40 nm. At the same time, the performance of these devices was comparable to devices with an evaporated electrode. Reducing the thickness of PEDOT:PSS layer from 1200 nm to 40 nm is a very efficient way to reduce the manufacturing costs of OPV devices. In addition, combining of thin PEDOT:PSS layer with an inkjet printed Ag grid structure allows manufacturing of semi-transparent devices.

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1. Introduction

Organic photovoltaics (OPVs) attract a lot of attention due to their low cost potential which can be achieved via all-solution roll-to-roll (R2R) processing [1–7]. Towards all-solution processing, a lot of effort has been put into the development of an alternative for ITO as a front electrode [8–16]. However, the back electrode also has to be produced from solution. PEDOT:PSS as a hole transport layer followed by the deposition of Ag is a typical back electrode in organic solar cells with the inverted configuration. Although it is well accepted that this electrode can be solution processed, often Ag is still deposited by thermal evaporation. Alternative solution processing deposition methods for Ag back electrodes are spray (aerosol) coating [7,17–20], inkjet printing [21–23], flexographic [21] or screen printing [21,24,25]. A substantial limitation in the solution processing of the back electrode is the requirement of

sintering at high temperature and under ambient conditions. These conditions can lead to degradation of the PEDOT:PSS and photo-active layers.

Screen-printing in ambient condition is the most explored so far in both lab scale devices and R2R production. While only limited sintering time was needed (3 min at 140 °C) to yield high conductivity, the majority of defects was caused by an interaction of the solvents from the screen print Ag ink with the underlying layers [22]. Moreover, screen printing is a contact method, which may also lead to an increasing number of defects in underlying layers. To protect underlying layers from aggressive effects of solvents and to avoid the mechanical stress from the screen printing process, the thickness of the PEDOT:PSS layer is significantly increased to provide a kind of 'buffer layer' protecting the photoactive layer [24,25].

Inkjet printing is a non-contact technique and might require thinner PEDOT:PSS layers, which is a positive aspect in terms of the final cost of OPV. Previously, an inkjet printed back electrode has been successfully demonstrated with a 800 nm thick PEDOT:PSS layer in organic solar cells with an inverted cell configuration [22].

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In this work, we further explore the type of PEDOT:PSS and the effect of its thickness over a large series of devices with inkjet printed back electrodes.

2. Experimental

Glass substrates of 30 mm × 30 mm covered with an ITO layer (sheet resistance 10 Ω/sq), patterned by photolithography, were obtained from Naranjo. Poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS, Agfa, S305) was provided by Agfa and used as received. Poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS, Agfa, EL-P5015) was provided by Agfa and diluted with isopropanol (IPA) in ratios of 1:1, 1:2, and 1:3. Poly(3-hexylthiophene) (P3HT, Plextronics Plexcore OS 2100) and [6,6] phenyl-C61-butyric acid methyl ester (PCBM, 99%, Solenne BV) were used as received. All solvents were acquired from Sigma-Aldrich and used as received.

Zinc oxide (ZnO) nanoparticles (diameter: 5–10 nm) were synthesized using a modified hydrothermal condensation process [26] and dispersed in acetone at a concentration of 10 mg/mL. Spin coating was done at 1000 rpm (acceleration 5000 rpm/s, with open lid) for 60 s. This resulted in a layer thickness of 20–40 nm. P3HT and PCBM in weight ratio of 1:1 were dissolved in a concentration of 26 mg/mL each in ortho-dichlorobenzene (oDCB). The solution was stirred for 16 h at 90 °C and filtered before usage. The solution was spin coated in air at 1000 rpm for 60 s (acceleration 1000 rpm/s, with open lid) and the resulting layer thickness was approximately 240 nm. Annealing of the photoactive layer was performed in a nitrogen environment at 130 °C for 10 min. PEDOT:PSS was spin coated in air with different speeds, resulting in a variety of layer thicknesses and annealed in a nitrogen environment at 130 °C for 10 min.

The Ag back electrode (100 nm) was thermally evaporated in a vacuum chamber through shadow masks at a base pressure below 2×10^{-6} mbar. For the inkjet-printed Ag electrodes a commercially available nano-particles (np) inks (Sun Chemicals, Slough UK) consisting of 20 wt% Ag (U5603) was used. The silver np ink was printed using a DMP-2800 materials printer (Dimatix-Fujifilm Inc., USA), equipped with a 10 pL cartridge (DMC-11610, Dimatix-Fujifilm Inc., USA). Printing was performed using a drop space of 20 μm, a voltage of 20 V, a print head temperature of 30 °C, a frequency of 10 kHz and a customized waveform. After printing, the samples were oven sintered for 10 min at 130 °C using a Memmert hot air oven. For the screen printed electrode the Toyochem Rexalpha RA FS FD 076 UV curable Ag ink was used. The ink was cured in a nitrogen environment upon irradiation with a Steuernagel lamp (575 W) for 1 min.

Surface topography, thickness and cross-sectional areas of the printed silver tracks were measured with an optical profilometer (Wyko NT9100, Veeco, Mannheim, Germany) and optical micrographs were taken using an optical microscope (Leika DM 2500M).

Current density–voltage (J – V) characteristics were measured in nitrogen with a Keithley 2400 source meter between -2 and $+2$ V using simulated AM 1.5 global solar irradiation (100 mW/cm²) with halogen lamp. The cell area, which is defined by the overlap between ITO and the full area Ag electrode, was 0.367 cm². Measurements were done with a mask to have well-defined illuminated areas of 0.25 cm². The grid electrode consists of three lines with a pitch size of 2 mm and the deposition width of 100 μm. The final width of the lines depended on the deposition method. Short-circuit currents and PCEs representing AM1.5G illumination conditions were estimated by integrating the EQE with the AM1.5G solar spectrum. All device fabrication steps were performed under CR1000 conditions.

3. Results and discussion

Printing of back electrodes in organic solar cells with inverted architecture is limited by some practical considerations. For example, chemical treatments or high temperature sintering cannot be applied, because it can lead to degradation of the underlying PEDOT:PSS and photoactive layer. Although sintering time is usually limited to few minutes, the main source of degradation is the solvents from the Ag ink formulation. In order to overcome this problem, the thickness of the PEDOT:PSS layer was increased up to 1 μm [22,24]. Here, the effect of PEDOT:PSS thickness and type of PEDOT:PSS are explored over a big series of devices, to clarify the origin of this effect.

3.1. Hole transport layer EL-P5015

EL-P5015 is a screen print formulation based on PEDOT:PSS to be used as ITO alternative or as a back electrode in an inverted OPV stack. It is a highly viscous, solvent based formulation. Hence, in some cases, the formulation is diluted with IPA to obtain a coatable, solvent based formulation [22,27,28].

A series of devices with following device stack has been produced: glass/ITO/ZnO/P3HT:PCBM/PEDOT:PSS/Ag. The thicknesses of PEDOT:PSS layers were 1200 nm, 600 nm, 300 nm and 200 nm. To obtain these thicknesses, the EL-P5015 PEDOT:PSS was diluted with IPA in the ratios of 1:1, 1:2 and 1:3 and spin coated with 1000 rpm, leading to 1200, 600 and 300 nm, respectively. In addition, the 1:3 dilution was also spin coated at 2000 rpm, resulting in a thickness of 200 nm. The silver back electrode was subsequently either thermally evaporated through a shadow mask, or inkjet printed (with full area coverage or as current collecting grid). The full area electrode was used for both evaporation and printing, while the grid electrode was applied only by printing.

The photovoltaic parameters of the devices with an evaporated Ag electrode are shown in Table 1. A small decrease of the fill factor (FF) is observed with decreasing PEDOT:PSS layer thickness, which probably can be attributed to the increase of the resistance in PEDOT:PSS layer. The short circuit current density in the devices with thinner PEDOT:PSS layer increases due to increasing transparency of PEDOT:PSS layer which leads to a higher reflection of light by the evaporated Ag. However, on average the changes in the device parameters do not lead to a significant change in performance for the range of devices with different thicknesses of PEDOT:PSS.

Inkjet printing of both full area and grid electrode on the devices with different PEDOT:PSS thicknesses shows a decreasing reproducibility of the device performance with decreasing PEDOT:PSS layer thickness. The average photovoltaic parameters of the devices with inkjet printed back electrode and different PEDOT:PSS thicknesses are shown in Table 1. Fig. 1 illustrates reproducibility of photovoltaic

Table 1

Average photovoltaic parameters of the devices with EL-P5015 PEDOT:PSS as hole transport material.

Ag electrode	Deposition method	Thickness [nm]	V_{oc} [V]	FF [%]	J_{sc} [mA/cm ²]	PCE [%]
Full area	evaporated	1200	0.540	63.0	8.82	3.00
		600	0.540	62.9	9.13	3.10
		300	0.540	60.6	9.47	3.09
Full area	inkjet	1200	0.527	58.8	8.53	2.64
		600	0.522	55.1	8.53	2.45
		300	0.519	54.7	8.48	2.41
		200	0.515	52.5	8.31	2.25
Grids	inkjet	1200	0.539	58.4	7.79	2.45
		600	0.525	52.1	7.74	2.11
		300	0.521	49.0	7.60	1.94
		200	0.518	41.0	7.58	1.61

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