

Contents lists available at ScienceDirect

# Solar Energy Materials & Solar Cells



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

# Hybrid electrohydrodynamic atomization of nanostructured silver top contact for inverted organic solar cells



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#### ARTICLE INFO

Article history: Received 14 March 2014 Received in revised form 24 June 2014 Accepted 25 June 2014

Keywords: Hybrid electrohydrodynamic atomization Silver top electrode Surface morphology Inverted organic solar cells Electrical study

### ABSTRACT

The present study is demonstrating the deposition of solution based nanostructured silver top electrode for inverted organic solar cells. The deposition of silver nanoparticles through hybrid electrohydrody-namic atomization (Hybrid-EHDA) is discussed in detail. The film thicknesses were varied with respect to spray times. The interface between the metal electrode and organic film was investigated using a focused ion beam analyzer. The sheet resistance of nanostructured silver film was reduced by increasing the film thickness. The lowest sheet resistance of silver film is ~0.07  $\Omega$ /sq with a film thickness of 2.6  $\mu$ m. The power conversion efficiency of fabricated devices was found to be influenced by the sheet resistance of silver film. The maximum power conversion efficiency (~2.44%) was observed at 0.07  $\Omega$ /sq. Our results demonstrate that the Hybrid-EHDA will play a promising role in the fabrication of inverted organic solar cells using a solution based top electrode.

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#### 1. Introduction

Nowadays, photovoltaic technologies are of considerable interest in the development of renewable energy as a replacement for the non-renewable technologies due to increasing energy demand and carbon dioxide (CO<sub>2</sub>) emission. Currently, the silicon based solar cells have become firmly entrenched within the photovoltaic (PV) market [1–3]. However, due to the high production cost and material scarcity of silicon-based solar cells, scientists and engineers are of much interest to the development of alternative materials for solar cell applications. In this aspect, organic polymeric materials have good advantages such as low cost, light weight, solution processability and its potential for the large area fabrication at low temperature [4-6]. However, bulk heterojunction based organic polymers are of significant interest in organic PV cells, which are obtained by the intercalation of optically active polymers and electron accepting molecules [7]. Organic photovoltaic (OPV) cells fabricated without encapsulation lead to the oxidization of respective metal electrodes and the degradation of active layers in the presence of air due to the influence of oxygen and moisture diffusion [8]. In addition, the deposited metal electrode may diffuse into the active layer, which leads to an

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*E-mail addresses: ikim@kimm.re.kr* (I. Kim), amm@jejunu.ac.kr (K.-H. Choi). <sup>1</sup> These authors contributed equally. alteration in the physico-chemical properties of the organic active layer. In order to mitigate this issue, introducing a buffer layer between the electrode and active layer to reduce the physicochemical degradation of the active layer and prevent the oxygen and moisture diffusion into the device [9,10]. The inverted structure of OPV cells significantly improves the device stability using metals less sensitive to the air such as silver (Ag) or gold (Au), as a hole collecting electrode, and indium tin oxide (ITO) as an electron receiver. Inverted organic solar cells were also encouraged by using poly(3,4-ethylenedioxythiophene):poly(styrenesulphonate) (PEDOT:PSS) as a hole transport buffer layer between the active layer and Ag electrode, which effectively reduces oxygen and metal diffusion into the active layer [10,11].

In the past decade, several groups have reported that solutionbased non-vacuum printing techniques such as screen printing, doctor blading, inkjet printing and spray coating can be used for the deposition of active layer and buffer layer in the fabrication of polymer solar cells [12–15]. Such efforts have mainly focused on the large-scale fabrication of organic layers, with the exception of the top electrode in organic solar cells. Whereas vacuum based techniques have been widely used for the metal top electrode [12,16,17], these techniques have suffered from several limitations including the use of complex processes, a controlled environment and expensive vacuum systems that may cause thermal damage to previously deposited organic layers [18,19]. In order to overcome the above problem, solution based techniques are of very much interest as an alternative technique to develop the top electrode

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using metal nanoparticle inks [20,21]. However, the solution based deposition of metal nanoparticles is a challenging task due to solvent influence on the underlying organic layers [20]. This issue can be mitigated by using one of several advanced printing techniques, such as hybrid electrohydrodynamic atomization (Hybrid-EHDA), which strictly reduces the solvent impact during device fabrication.

The Hybrid-EHDA is the most promising and suitable technique for patterns and thin film fabrication in coating industries [22,23]. This technique offers low cost, high processing speed, less material wastage, large area printing within a quick time period [23,24]. The basic principle of the Hybrid-EHDA system is the atomization of starting precursors or inks under the influence of electric potential and pressure to overcome the liquid surface tension and produce surface charged droplets with a diameter ranging from sub-micron to nano-scale. A random distribution of tiny droplets is deposited on the target substrates. The significant advantage of this technique is the ability to cover a large area with fast drying droplets in the order of microseconds. This acts to control the solvent impact on the underlying organic layers and requires a very low temperature for annealing, as compared to other solution-based process [20,23].

In this study, we investigate the deposition of nanostructured silver through solution based Hybrid-EHDA. The physico-chemical properties of fabricated organic solar cells using a silver top electrode with different film thicknesses are examined through field-emission scanning electron microscopy (FE-SEM), atomic force microscopy (AFM), focused ion beam (FIB) and current-voltage (J–V) analysis.

#### 2. Experimental procedure

#### 2.1. Hybrid electrohydrodynamic atomization (Hybrid-EHDA) setup

The block diagram and the experimental setup of Hybrid-EHDA are represented in Figs. 1a and S1. Silver ink (30 wt%) was filled in disposable plastic syringe and the ink was driven by a piston pump. The syringe was directly connected to a capillary nozzle holder. A metallic nozzle with an internal diameter of 150  $\mu$ m was used for the atomization process and the nozzle was fixed at the bottom of the nozzle holder. The potential was applied in between the metallic nozzle and the substrate through a high voltage power source (0–21 kV). The substrate holder was ground. The applied potential was used to control the atomization process,

which was further controlled by a pneumatic pressure pump (0– 5 bar). The movement speed of the substrate and nozzle holder was controlled via the x-, y- and z- axes. The overall experiment was monitored through a CCD camera, which was interfaced with PC.

#### 2.2. Fabrication of organic solar cells

The organic device (ZnO/P3HT:PCBM/PEDOT:PSS/Ag) was fabricated on a patterned ITO glass substrate (Merck Display Technologies, 15  $\Omega$ /sq,) according to the following procedure.

# 2.2.1. Deposition of ZnO on ITO coated glass substrate

Zinc oxide ink was prepared by the uniform dispersion of ZnO nanoparticles (1 g) in 2-methoxyethanol and the ink was stabilized using N-methyl ethanolamine. The procedure was as detailed in the literature [25]. Freshly prepared ZnO was used to deposit the ITO coated glass substrate via spin coating [26]. Before the deposition process, the substrates were cleaned using acetone, isopropanol and deionized water followed by UV irradiation using a UV cleaner for 10 min. The deposited of ZnO thin film was achieved at a rotational speed of 1000 rpm for 30 s under ambient conditions and it was annealed at 150 °C for 2 h.

## 2.2.2. Deposition of P3HT:PCBM (active layer)

Freshly prepared composite P3HT:PCBM (1:1) ink was spin coated on ZnO/ITO-glass substrate from o-dichlorobenzene solution and the P3HT:PCBM ink preparation was as reported in the literature [27,28]. The homogeneous deposition of P3HT:PCBM thin film was achieved at a rotational speed of 1000 rpm for 30 s and the obtained film was annealed at 120 °C for 40 min under N<sub>2</sub> atm.

#### 2.2.3. Fabrication of PEDOT:PSS as a buffer layer

The PEDOT:PSS aqueous solution was used as a buffer layer in organic solar cells. The hydrophobic surface of the pre-deposited P3HT:PCBM (active layer) was reduced with oxygen ( $O_2$ ) plasma treatment and exhibited hydrophilic nature [20]. Subsequently, the PEDOT:PSS film was deposited on the active layer through spin coating. The PEDOT:PSS thin film was achieved at 300 rpm for 30 s. The deposited PEDOT:PSS layer was annealed at 140 °C for 10 min.

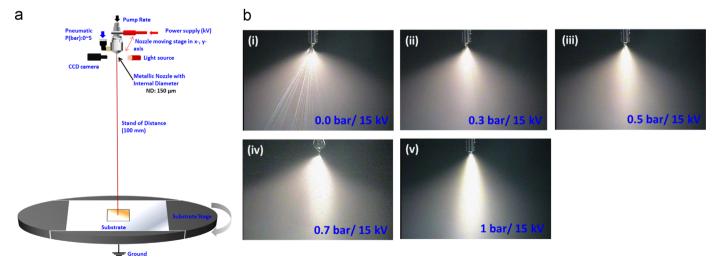


Fig. 1. (a) Schematic representation of hybrid electrohydrodynamic atomization, and (b) the stable spray mode or atomization mode of silver ink at a constant applied potential (15 kV) with different pneumatic pressures.

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