Solar Energy Materials & Solar Cells 125 (2014) 72–77

Contents lists available at ScienceDirect

Solar Energy Materials & Solar Cells

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

A study of the effects of drying and sintering on the performance of organic photovoltaic devices

Joong Hyun Yeom [a], Manho Kim [a], Sehun Rhee [a,*], Jae-Woong Yu [b,**]

[a] Department of Mechanical Engineering, Hanyang University, 17 Haengdang-dong, Seongdong-gu, Seoul 133-791, Republic of Korea
[b] Department of Advanced Materials Engineering for Information & Electronics, Kyung Hee University, 1732 Deogyeong-daro, Giheung-gu, Yongin, Gyeonggi 446-701, Republic of Korea

ARTICLE INFO

Article history:
Received 8 August 2013
Received in revised form
10 February 2014
Accepted 20 February 2014
Available online 12 March 2014

Keywords:
Organic photovoltaic
Processing
PEDOT annealing
Silver sintering
Far-infrared oven
Microwave oven

ABSTRACT

The effects of different processing methods on the performance of organic photovoltaics (OPVs) for the hole extraction layer and printed metal layers were studied. Four different methods (hot plate, air oven, vacuum oven and far-infrared oven) were utilized to dry hole extraction layers formed from aqueous nano-particle dispersions. The far-infrared oven showed the best drying characteristics and the sheet resistance was 69 Ω/□ in 240 s. To sinter a screen printed silver electrode, six different methods (hot plate, air oven, vacuum oven, far-infrared oven, UV lamp and microwave oven) were employed. The microwave oven yielded the most effective sheet resistance drop, as low as 0.06 Ω/□ in 20 s when the power of the microwave oven was 400 W. Using the fast processing methods developed in this study, the processing time can be reduced by about 40% without a loss in the performance of prepared devices.

1. Introduction

Organic photovoltaic (OPV) devices possess several advantages over traditional silicon-based or other inorganic solar cells, due to their light weight, low cost, solution processability, and the possibility of custom tailoring the structure. Consequently, OPVs have been investigated as a new way of obtaining clean and inexpensive renewable energy. Research and development of electronic devices based on organic materials began some time ago, leading to today's much more intensive and continuous studies into advanced flexible devices. In particular, the mechanical flexibility of polymeric materials and the capability of solution processing allow OPVs to be naturally compatible with printing methods through the use of plastic substrates. Complicated device preparation methods, including photolithography and vacuum processes can be easily replaced by printing methods when an appropriate polymeric material is used to fabricate devices.

The implementation of high throughput roll-to-roll printing techniques into flexible electronics processing is essential for the price compatibility of OPVs, as their performance is low when compared to their inorganic counter parts. Cost analyses of OPV devices for roll-to-roll processing have been reported in the literature [1,2]. Previous research suggests that it is critically important to adopt the high speed device fabrication method. Since roll-to-roll techniques are based on solution processing, it is important to adopt appropriate protocols (i.e., drying, annealing and sintering) for fast device fabrication. According to many roll-to-roll OPV printing studies [3–12], the drying unit in a roll-to-roll system should be much larger than the coating system for proper device fabrication [4]. Krebs et al. [5] reported on the processing times (total processing, drying and waiting time) required for long webs (200 m long).

In the case of inverted type OPVs, a typical device is composed of an electron extraction layer (typically composed of either TiO_2 or ZnO nano-particles) on top of a substrate, an active layer (usually a polymer donor/fullerene derivative acceptor blend), a hole extraction layer [typically thick poly(3,4-ethylenedioxythiophene) (PEDOT) layer] and a printed metal layer (e.g., silver) as a collector. The active layer formed by a polymer donor and fullerene acceptor blend shows phase separation which strongly depends on the annealing conditions [11]. While slow annealing leads to vertical and lateral phase separation, fast drying leads to a larger P3HT crystalline structure and an increased amount of PCBM on the top of the film surface [11]. Since this active layer is an immiscible blend system, an appropriate annealing time should be used to optimize the morphology and increase device efficiency. Compared to the other constituent layers, the thick

* Corresponding author.
** Corresponding author.
E-mail addresses: srhee@hanyang.ac.kr (S. Rhee), jwyu@khu.ac.kr (J.-W. Yu).

http://dx.doi.org/10.1016/j.solmat.2014.02.030
0927-0248

PEDOT layer, which is formed from an aqueous dispersion solution, requires a long drying distance or low drying speed [9]. Finally, the printed metal collector, which is usually formed from a paste ink of dispersed silver nanoparticles or microflakes, also requires a long sintering time for optimum resistivity. In other words, a reduction in the time required for the entire drying process is very important for price competitiveness. Such a scenario can be realized by optimizing the processing conditions for the hole extraction and metal collector layers. Some researchers have employed a single drying/annealing procedure (e.g., microwave [13,14], IR [15]) for the fabrication of OPV devices. Typical inverted type OPV device fabrication requires the processing of four different characteristic layers: electron transporting nano-particles dispersed in alcohol, a polymer blend in an organic solvent, hole conducting nano-particles dispersed in an aqueous solution, and metal nanoparticles or microflakes dispersed in an organic solvent. Thus, it is very important to establish a proper processing method for each layer in order to develop a high throughput production method such as roll-to-roll processing. In this study, we examined the effects of different drying/sintering techniques on the performance of OPVs with PEDOT and printed silver layers.

2. Materials and methods

2.1. Fabrication of devices

An inverted type OPV cell was prepared. A typical organic solar cell used for the active layer treatment studies was fabricated as follows. A patterned ITO cell ($<15\ \Omega/\square$ on glass and $<30\ \Omega/\square$ on PEN) with an insulator was prepared by photolithography and cleaned with acetone, isopropyl alcohol, and deionized water. The clean ITO was then treated with UV-Ozone for better coating characteristics. TiO_2 nanoparticles were synthesized according to the previously reported method [16,17]. The TiO_2 film, which is used as an electron extraction layer, was spin coated from a methanol solution (3 mg/mL concentration) and dried for 10 min at 110 °C. For the active layer, 4 wt% of regioregular Poly[3–hexylthiophene–2, 5–diyl] (P3HT) (purchased from Aldrich) and PCBM (purchased from Nano-C) blended in a 1: 0.6 weight ratio was dissolved in anhydrous chlorobenzene and spin-cast onto ITO coated with a homemade TiO_2 nanoparticle layer. The active layer, which had a thickness of 400 nm, was dried and annealed in a far-infrared radiation heater (peak radiation frequency: 3.5–4 μm, maximum surface temperature of 680 °C, power of 600 W, Misumi) at ambient conditions. The hole extraction layer of poly(3,4-ethylenedioxythiophene): poly(4-styrene sulfonate) [PEDOT: PSS] (Agfa E5010) was blade cast on top of the active layer and dried for 10 min at 110 °C using four different annealing methods. Finally silver paste (PARU, MicroPE®PC-010, 200 cP, particle size: 20 nm), which acts as a positive electrode, was screen printed through a patterned polyimide film screen. The thicknesses of each layer, as determined by cross-sectional TEM analysis, were 0.2, 0.4, 5 and 10 μm for the TiO_2, active layer, PEDOT, and silver electrode, respectively. Fig. 1 shows the structure of the fabricated OPV devices. For an analysis of the silver electrode sintering characteristics, a far-infrared heater, hot plate, air oven, vacuum oven, 3 types of UV lamps (mercury UV, metal halide UV, and gallium UV) and a commercial household microwave oven (in-house modified for research) were employed. The prepared device was encapsulated with a glass plate using UV curable resin.

2.2. Characterization

The absorption spectra of the films were obtained with a photodiode array type UV–vis spectrometer (HP 8453). Sheet resistance was measured via the four probe method with a Keithley 2600. An Oriel Class A type solar simulator (IEC 904) with an Oriel Reference Cell (reference cell has been calibrated with NREL data) was used as a light source, and all measurements were performed under a 1 sun condition (AM1.5 100 mW/cm^2). The *J–V* characteristics were measured with a Keithley 2400 source-measure unit; the data were not corrected for reflection losses and light absorption in the ITO electrode.

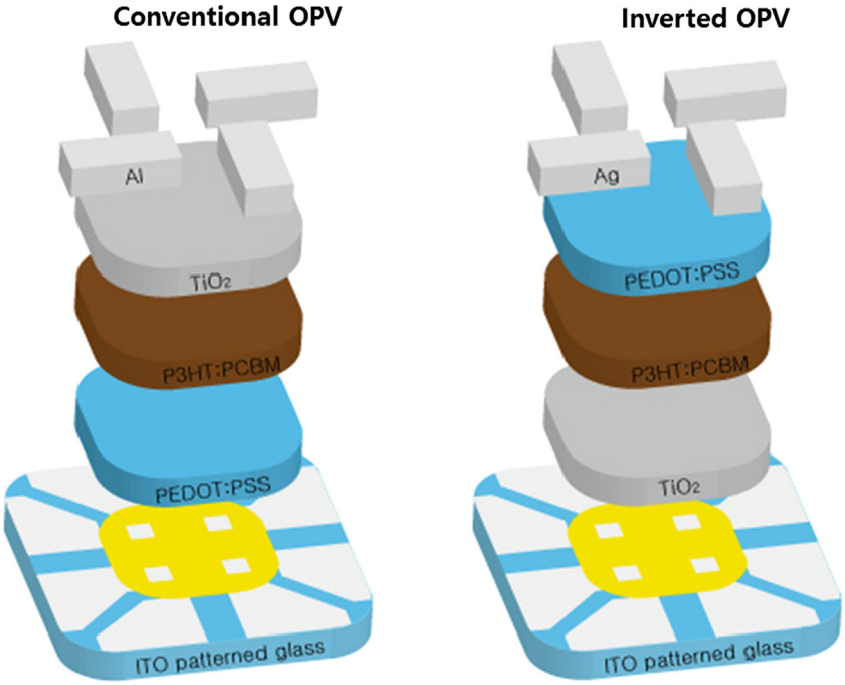

Fig. 1. Schematic diagrams of the OPV device structure used in this study.