Thin Solid Films 567 (2014) 1–7

Contents lists available at ScienceDirect

Thin Solid Films

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

Performance improvement of inverted organic solar cells by adding ultrathin Al_2O_3 as an electron selective layer and a plasma enhanced chemical vapor deposition of SiO_x encapsulating layer

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ARTICLE INFO

Article history:
Received 2 March 2014
Received in revised form 18 July 2014
Accepted 22 July 2014
Available online 2 August 2014

Keywords:
Atomic layer deposition
Aluminum oxide
Inverted organic solar cells
Plasma-enhanced chemical vapor deposition
Encapsulation

ABSTRACT

In this paper, we report the performance improvement of inverted organic solar cells by adding an ultrathin electron selective layer of Al_2O_3 prepared between the indium tin oxide (ITO) electrode and the active transport layer through atomic layer deposition (ALD). We evaluated the cell shelf-life after encapsulating with SiO_x-coated polyethylene terephthalate, where the SiO_x layer was made by plasma enhanced chemical vapor deposition (PECVD). It was found that the devices with ALD Al_2O_3 have a higher open circuit voltage than those without the ALD Al_2O_3 layer. Al_2O_3 deposited on an ITO electrode decreased the work function of ITO. Furthermore, based on the current density–voltage curves of the initial devices showing a pronounced S-shape, we soaked the cells with the ultraviolet (UV) light process. Then we obtained a higher efficiency in these ALD Al_2O_3 treated devices. With a careful analysis by atomic force microscopic and X-ray photoelectron spectroscopy, we believe that the UV light soaking process affected both ITO and Al_2O_3. Further, after the encapsulation by PECVD SiO_x, our devices achieved a shelf-life of over 500 h for 50% retained cell efficiency.

1. Introduction

As a class of renewable clear energy resources, organic solar cells (OSCs) have attracted much attention in recent years. OSCs are potential energy resources in the solar energy harvesting industry because they are flexible, low-cost, light weight and scalable [1,2]. The typical structure of conventional OSCs consists of indium tin oxide (ITO)/poly(3, 4-ethylene dioxythiophene doped with polystyrene sulfonate) (PEDOT:PSS)/poly-3 hexyl-thiophene-2, 5-diyl (P3HT): 6, 6-phenyl-C61-butyric acid methyl ester (PCBM)/metal electrode. The metal electrode should have a low work function in general [3]. However, the electrode with a low work function in OSCs is susceptible to degradation by oxidization when exposed to ambient environment, as the oxide layer formed is insulating, which can reduce the electronic conductivity. Additionally, the structure of the ITO/PEDOT:PSS interface is not stable for cell assembling, since the PEDOT:PSS can chemically erode the ITO electrode. Both of the above problems can degrade the performance of OSCs and lead to a poor stability of OSCs in operation [4–6]. Therefore, the strategic structures of inverted and encapsulated OSCs were proposed, where, typically, a metal oxide such as TiO_2, ZnO_2 is used as an electron selective layer on an ITO cathode and an air-stable high-work-function metal is used as the top anode [7–10].

In this paper, we utilized atomic layer deposition (ALD) to prepare an ultra-thin Al_2O_3 film as the electron selective layer in the inverted OSCs, since ALD is especially suitable in fabricating highly uniform, dense, conformal and ultra-thin films with their film thicknesses being able to be precisely controllable [11]. We firstly studied the role of Al_2O_3 as an electron selective layer on the photovoltaic conversion efficiency. Then, polyethylene terephthalate (PET) films with SiO_x coating through plasma enhanced chemical vapor deposition (PECVD) were utilized to encapsulate the inverted OSCs for improved stability. Finally the shelf-life of encapsulate inverted OSCs was evaluated.

2. Experimental methods

Patterned ITO glass substrates (ITO thickness is 180 nm) were ultrasonically cleaned in detergent, alcohol, acetone and alcohol sequentially for 10 min, respectively. Then the substrates were dried by N_2 stream and treated with ultraviolet (UV)–ozone for 5 min. 5 cycles of ALD Al_2O_3 were then deposited on the cleaned ITO substrates at 110 °C. During the Al_2O_3 ALD process, the dosing times of the precursors of trimethyl aluminum (TMA) and water were both set at 0.05 s, and the N_2 purging times (100 sccm) were set at 30 s and 60 s for TMA and water, respectively. Then the Al_2O_3 coated ITO substrates were transferred into a nitrogen atmosphere glove box for sequential spin-coating of 1 wt.% P3HT and 0.8 wt.% PCBM in chlorobenzene (27 mg/mL) at 800 rpm for 30 s. Then the samples were baked at 110 °C for 10 min. Finally, a MoO_3 layer (10 nm) and a Ag layer

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http://dx.doi.org/10.1016/j.tsf.2014.07.038
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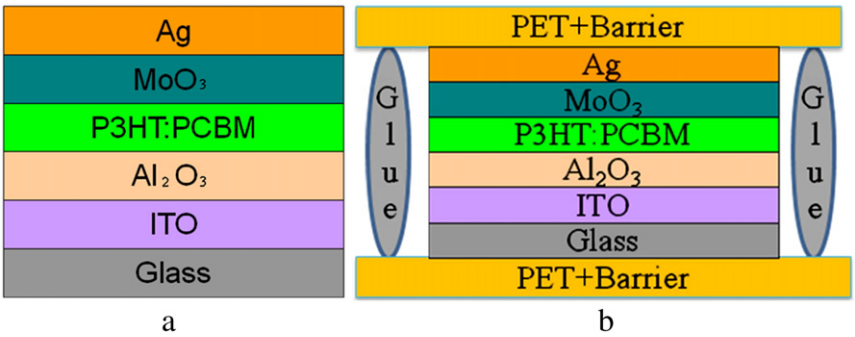

Fig. 1. Schematic structure of a — inverted organic solar cells; b — encapsulated inverted organic solar cells.

(100 nm) were sequentially deposited through a shadow mask to define an active area of 4 mm^2, respectively. Fig. 1(a) illustrates the schematic structure of our devices.

An encapsulated device of inverted OSCs consists of polyethylene terephthalate (PET) + barrier coating/ITO anode/Al_2O_3/P3HT:PCBM/MoO_3/Ag cathode/PET + barrier coating as shown in Fig. 1(b). The barrier layer used in this study was a 125 μm transparent PET substrate coated with low permeable SiO_x layers by PECVD on both sides. The gap between the two barrier coatings was sealed by epoxy glue. The PECVD process parameters of SiO_x deposition on both sides of the PET substrates was the identical. We used 40 kHz pulsed power to ignite the plasma, with hexamethyldisiloxane (HMDSO) (98%) as the precursor, oxygen (O_2) (99.999% in purity) as the oxidant gas, and argon (99.99% in purity) as the diluting and ionizing gas. The schematic diagram of the plasma setup for SiOx deposition is shown in Fig. 2 [12]. In comparison, except for the encapsulating process, the non-encapsulated cells were prepared with the same process, and all of the solar cell devices were stored and measured under the same conditions.

The characteristic current density–voltage (J–V) curves of the OSCs were measured by a Keithley 4200 under a xenon lamp (100 mW/cm^2). The light intensity from the light source was calibrated previously by a standard silicon based solar cell. Each data was averaged from 5 samples. The barrier properties of the SiOx-coated PET were characterized by oxygen transmission rate (OTR) and water vapor transmission rate (WVTR) with the Mocon instrument OX-TRAN Model 2121, which has a sensitivity of 5×10^{-2} $cm^3/(m^2 \cdot day)$ at 23 °C and 0% relative humidity, and AQUATRAN Model 1, which has a sensitivity of 5×10^{-4} $g/(m^2 \cdot day)$ at 37 °C and 100% relative humidity, respectively. The light transmission rate was gained by a UV–visible spectrophotometer (UV-2501 pc). Surface topographies of ITO and ITO/Al_2O_3 were analyzed by an atomic force microscope (AFM) (Veeco, USA) in the tapping-mode, and the average roughness (Ra) was obtained in a range of a 1.0×1.0 μm^2 scan. Chemical state of ITO/Al_2O_3 was characterized by X-ray photoelectron spectroscopy (XPS, Kratos Axis ULTRA, using a monochromatic Al $K\alpha$ source = 1486.7 eV). All XPS spectra reported here were referenced to C1s peak (284.8 eV). The film thickness was measured by spectroscopic ellipsometry (SE, Horiba) at a fixed angle (70°) from 190 nm to 826 nm.

3. Results and discussion

3.1. The role of Al_2O_3 as an electron selective layer in inverted OSCs

Firstly, we investigated the role of Al_2O_3 as an electron selective layer in the inverted OSCs. Al_2O_3 as an electron selective layer was prepared by ALD technique for 5 cycles at 110 °C. The thickness of Al_2O_3 was estimated as 0.62 nm based on the deposition rate of ca. 0.124 nm/cycle. This deposition rate was measured by SE on the 100- and 250-cycled ALD Al_2O_3 samples, which is consistent with previous studies [13]. Fig. 3 and Table 1 show the J–V curves and device parameters with/without ALD Al_2O_3, respectively. Both devices show a pronounced S-shaped J–V curve, and the 5 cycles of ALD Al_2O_3 treated device even exhibits a lower fill factor (FF) and therefore a lower photovoltaic conversion efficiency (PCE).

As FF is determined by the charged carriers reaching the electrodes [14], there is a competition between charged carrier recombination and transport. A smaller FF in the device with 5 cycles of ALD Al_2O_3 must be related to the weaker electrical transport in insulative

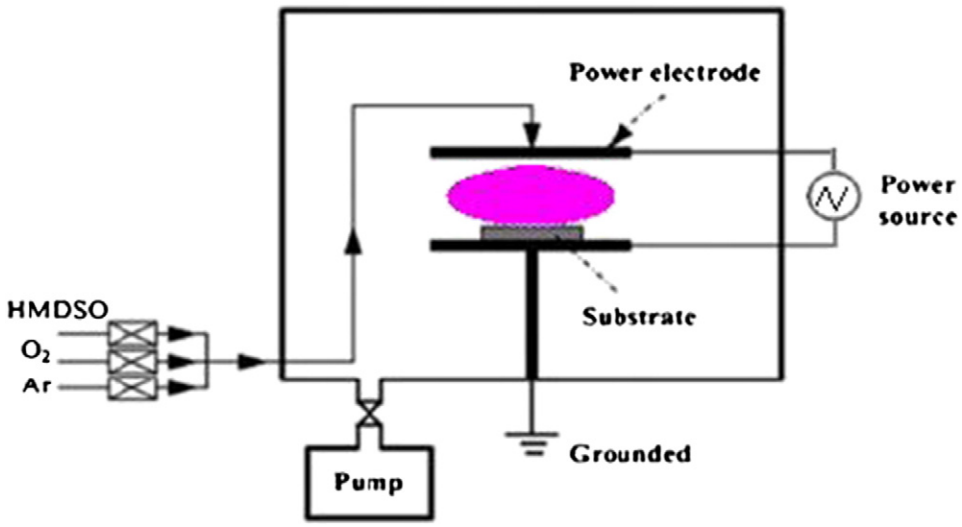

Fig. 2. Schematic diagram of plasma setup for PECVD SiO_x.