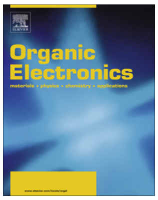
Organic Electronics 19 (2015) 140–146

Contents lists available at ScienceDirect

Organic Electronics

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



Stable organic photovoltaic with PEDOT:PSS and MoO_X mixture anode interfacial layer without encapsulation

Seung Joo Lee [1], Byung Soon Kim [1], Jae-Yeon Kim [1], Abd. Rashid bin Mohd Yusoff [1], Jin Jang *

Department of Information, Display and Advanced Display Research Center, Kyung Hee University, Dongdaemun-ku, Seoul 130-171, Republic of Korea

ARTICLE INFO

Article history:
Received 8 August 2014
Received in revised form 6 January 2015
Accepted 20 January 2015
Available online 28 January 2015

Keywords:
Organic photovoltaic
PEDOT:PSS
Metal oxide
Stable

ABSTRACT

Herein, we report about an efficient and stable organic photovoltaic that uses a poly(3,4-ethylenedioxythiophene):poly(styrene-sulfonate) (PEDOT:PSS) and molybdenum oxide (MoO_X) mixture for the anode interfacial layer, and that can reach 4.43% power conversion efficiency (PCE) under AM1.5 conditions. Utilizing PEDOT:PSS:MoO_X (1:1), the shelf life-time of poly(3-hexylthiophene) (P3HT), and indene-C_{60} bisadduct (ICBA)-based solar cells without encapsulation, can be realized with only a 25% deterioration after 672 h of storage in air. Furthermore, we compare the photovoltaic performance of the P3HT:ICBA-based organic photovoltaic with PEDOT:PSS, and PEDOT:PSS:MoO_X, in which PEDOT:PSS:MoO_X has outperformed the others. In addition, the water vapor transmission rate of PEDOT:PSS:MoO_X is 0.17 gm/(m^2 day), which is much less than that of PEDOT:PSS.

1. Introduction

Organic photovoltaics (OPVs) now receive more attention because of various factors, such as being lightweight, easy to fabricate, and cost effective [1–8]. Today, we have seen rapid improvement in terms of power conversion efficiency (PCE), and device lifetime [9]. The interface engineering between electrodes, interface, and photoactive layers remarkably influence the devices performance, including the fill factor (FF), short-circuit current density (J_{sc}), open circuit voltage (V_{oc}), stability and lifetime [10]. Hence, copious amounts of work for interface engineering has been put forward to achieve high performance and stable OPVs [5–8]. Notably, a few issues must be taken into account when considering an interfacial layer including transparency, conductivity, morphology, stability, and processability [10,11].

In most organic devices, poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) is frequently used as an anode interfacial layer for improving anode contact and also used to increase hole collection. Nevertheless, it is highly acidic nature possess a major drawback, where it can easily etch the commonly used indium tin oxide (ITO). Although this phenomenon usually deteriorates device performance and is less stable [12], uses alternative methods, such as utilizing metal oxide materials [13].

Solution-processed molybdenum oxide (MoO_X) has gained more attention as a potential anode interfacial layer [14–22], due to its hole mobility, environmental stability, and high transparency in the visible regime. According to Meyer and co-workers, the spin-coated MoO_X layer led to an excessive root-mean-square (rms) roughness; therefore, a thicker layer was required in order to avoid any leakage in their devices. Then, a sol–gel method was introduced to develop a solution processed MoO_X. However, this method is less suitable for the roll-to-roll process because it requires a high temperature (>250 °C) to crystallize the MoO_X layer [15,16]. Hammond and co-workers developed a continuous MoO_X [17]; however, the MoO_X solution

---

* Corresponding author. Tel./fax: +82 2 961 0270.
E-mail address: jjang@khu.ac.kr (J. Jang).

[1] Tel./fax: +82 2 961 0270.

http://dx.doi.org/10.1016/j.orgel.2015.01.028
1566-1199/

needs O_2 plasma treatment, and its precursor is relatively oxygen-sensitive making it less suitable for large scale manufacturing. Liu and co-workers proposed another method for preparing MoO_X, where ammonium molybdate was used as a precursor and resulted in large MoO_X crystals instead of a smooth thin film. By introducing a low thermal annealing process, Yang and co-workers overcame the challenges faced with large MoO_X crystals [19]. In 2012, Zilberberg, and Jasieniak, and their respective co-workers developed a sol–gel processed MoO_X using a novel precursor [21,22]. Although they were able to obtain a successful smooth layer, it must be maintained in ambient conditions in air for a long time in order to hydrolyze and age the sol–gel solution. These procedures are time-consuming and incompatible with the high throughput and scale-up technologies. Recent development of simple and time-saving MoO_X has been demonstrated by Tan et al., in which they have successfully presented highly efficiently OPVs with a PCE of 6.57% based on poly(3-hexylthiophene): indene-C_{60} bisadduct (P3HT:IC_{60}BA) bulk heterojunction (BHJ) system [23].

Therefore, in this letter, we demonstrate a simple, easy, and time-saving procedure to prepare a continuous and uniform MoO_X layer that can later be mixed with PEDOT:PSS. The MoO_X film was prepared by spin coating a $MoO_2(acac)_2$ isopropanol solution on pre-cleaned ITO glass, and then baking it in air at 150 °C for 10 min. The preparation procedure of the s-MoO_X layer is extremely simple and cost-effective; it employs a cheap and commercially available precursor, isopropanol, as a solvent and uses a low temperature process under ambient conditions, in which no additional sol–gel synthesis is needed. The obtained s-MoO_X layer is highly transparent in visible range and exhibits effective hole collection properties. To investigate the general suitability of the PEDOT:PSS:MoO_X anode interfacial layer in OPVs, a standard poly(3-hexylthiophene) (P3HT) was selected as an electron donor and a fullerene derivative indene-C_{60} bisadduct (IC_{60}BA) was chosen as the electron acceptors.

2. Experimental section

2.1. Device fabrication

According to the reported procedure, the aqueous MoO_X solution was prepared by a hydration method [22,24]. To prepare the MoO_X solution, ammonium heptamolybdate ($(NH4)_6Mo_7O_{24}–4H_2O$–Aldrich, 99.98%, 0.4 g) was heated in distilled water (10 mL) at 80 °C for 1 h. Using distilled water the MoO_X solution was distilled to the desired concentration. Afterwards, 5% of isopropyl alcohol was added into the aqueous MoO_X solution and stirred for 2 h at room temperature. The devices were based on the P3HT:ICBA active layer and the PEDOT:PSS:MoO_X (1:5) buffer layer. Afterwards they were processed in ambient atmosphere, with the exception of the thermal treatment in the glove box. Pre structured indium tin oxide (ITO) coated glass substrates were cleaned in acetone and isopropyl alcohol. After drying, the substrates are treated under the UV–ozone and, then coated with the LZO electron transport layer. After that, the samples were baked on a hot plate at 140 °C for 20 min. The P3HT:ICBA (1:1w/w) blended in dichlorobenzene (DCB) solvents were deposited on-to substrates using a spin-casting process. All devices are annealed at 150 °C for 10 min on a hot plate in a glove box. Later, the blend of PEDOT:PSS and MoO_X was used for the hole extraction layer (HEL) and was deposited on the photoactive layer. As a reference, PEDOT:PSS (40 nm) were also deposited. Finally, 100 nm Ag was evaporated through a shadow mask under 10^{-7} Torr. In this study, we have fabricated more than 16 devices.

The water vapor transmission rate (WVTR) value of the PEDOT:PSS and PEDOT:PSS:MoO_X films were measured at room temperature with 100% relative humidity. The measurements were carried out using PERMATRAN-W 3/33 (Mocon Inc., Minneapolis, USA). The films were cut into a uniform hexagon shape with a template and placed inside the text cells. The effective area of WVTR was 50 cm^2. The concentration of the water vapor was held at a different level by rotating the relative humidity pressure regulator. The system was purged using flowing a rapid stream of flowing nitrogen and then the sample was conditioned. In an actual run, the water vapor was introduced into the outer cell. The water vapor, diffused into the inner cell through the film was carried by a nitrogen stream to the infrared detector. The detector shows a linear response to the concentration of the water vapor in the carrier gas. The resulting signal which was calibrated using standard films was continuously recorded.

Surface morphologies of the photoactive layers were obtained by tapping mode using an atomic force microscope (AFM, Digital Instrument Multimode equipped with a nanoscope IIIa controller) and a scanning electron microscope (Hitachi S-4700).

XPS measurements were taken using a PHI 5000 VersaProbe (Ulvac-PHI) with a background pressure of 6.7×10^{-8} Pa, using amonochromatized Al Kα ($h\nu$ – 1486.6 eV) anode (25 W, 15 kV).

2.2. Device measurements

Current density–voltage ($J–V$) characteristics of the devices were measured using a Keithley 236 Source Measure Unit. Solar cell performance used a Newport Air Mass 1.5 Global (AM 1.5G) full spectrum solar simulator with an irradiation intensity of 100 mW/cm^2. $J–V$ characteristics of all devices were taken using a semiconductor characterization system (Keithley 2400 LV Source Meter under s simulated AM1.5 G spectrum with MAX-302, Asahi Spectra Co. Ltd., Japan). The EQE measurements were gathered using an EQE system (Model 74000) from the Newport Oriel Instruments USA and HAMAMATSU calibrated silicon cell photodiode used as a reference diode. The wavelength was controlled with a monochromator 200–1600 nm. All measurements were carried out at a room temperature of about 27 °C under a relative humidity of 60–70%.

3. Results and discussion

Further evidence of the purity and composition of the PEDOT:PSS and molybdenum oxide films were obtained