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Investigation into the effect of post-annealing on inverted polymer solar cells

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ABSTRACT

The work-function of indium tin oxide (ITO) electrodes was tuned with an interfacial dipole layer (WPF-oxy-F) to reverse the polarity in polymer solar cells (PSCs) with an inverted structure. The photoactive layer was based on poly(3-hexylthiophene) (P3HT) and [6,6]-phenyl-C61-butyric acid methyl ester (PCBM). Silver (Ag) and poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) were used as the top anode. The optimized conditions for the fabricated I-PSCs included an open-circuit voltage (V_{oc}) of 0.68 V, a fill factor (FF) of 64%, and a power conversion efficiency (PCE) of 3.86% through post-annealing at 170 °C. The high performance of I-PSC is due partly to the improved interfacial contact at active/PEDOT:PSS and mainly to the increase of the work-function of annealed PEDOT:PSS/Ag at 170 °C for 30 min. Here, we investigated the effect of post-annealing on I-PSC devices by carrying out various annealing sequences.

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1. Introduction

Recently, PSCs have attracted much attention due to their potential for easy fabrication by low-cost solution processing onto large areas of flexible substrates [1–3]. Conventional PSC devices that are based on P3HT:PCBM sandwiched by low work-function metal cathodes and PEDOT:PSS on ITO have achieved a PCE of ca. 4–5% via post-annealing treatment by thermal annealing [4]. However, low work-function metal still requires high-vacuum deposition, and it is easily oxidized under ambient conditions, which leads to the degradation of device performance [5].

To overcome these drawbacks, an inverted device configuration is used by reversing the polarity of charge collection in conventional PSCs. In general, higher work-function electrodes (e.g., Au, Ag and Cu) are less oxidized under ambient conditions and offer the possibility for printing and coating methods [5–7]. Transition metal oxides (MoO_3, WO_3,V_2O_5) and a conducting polymer (PEDOT:PSS) deposited on the active layer have been used for hole collection [8–11]. Of these, the combination of Ag and an adjacent PEDOT:PSS layer can substitute for air-sensitive Al as the anodic electrode for efficient hole collection [11]. In addition, inorganic metal oxides (Cs_2CO_3, TiO_2 and ZnO) have been used for electron collection by modifying the work-function of ITO [8–13]. However, the surface of metal oxides is known to have hydroxyl groups that cause charge trapping at the interface, leading to an increase in the resistance of PSCs [14,15].

In previous work, we showed that use of WPF-oxy-F and WPF-6-oxy-F, as conjugated polyelectrolyte-based interfacial layers [16–18], significantly improved the performance of both I-PSCs and normal PSCs [19–22]. These interfacial materials have several advantages: a conjugated backbone for good conductivity, ammonium salt and ethylene oxide side groups for large interfacial dipole tuning work-function of metals, and solubility in only polar solvents to prevent the intermixing of the upper layer. Our group recently reported that WPF-oxy-F reverses the polarity in I-PSCs with the Cu electrode as a bottom cathode [22]. These I-PSCs with WPF-oxy-F or WPF-6-oxy-F as a cathode interfacial layer and PEDOT:PSS/Ag as the anode were optimized through post-annealing treatment. Some I-PSCs reported by other groups also required post-annealing treatment [23,24]. However, to the best of our knowledge, the post-annealing effect in I-PSCs with polymer based interfacial layers [16–18,24,27,28] at both the cathode and anode electrodes has not been studied systematically, and therefore, it is poorly understood which layer or interface is more strongly affected by post-annealing treatment.

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Table 1
Summary of annealing conditions prior to Ag electrode deposition and after Ag electrode deposition (the shaded area indicates layers affected by annealing conditions).

Layers	Annealing conditions			
	(1)	**(2)**	**(3)**	Post-annealing
WPF-oxy-F (3 nm)	From 110 to 170 °C for 30 min	–	–	–
P3HT:PCBM (60 nm)	110 °C for 10 min	From 110 to 170 °C for 30 min	110 °C for 10 min	110 °C for 10 min
PEDOT:PSS (16 nm)	150 °C for 10 min	150 °C for 10 min	From 110 to 170 °C for 30 min	150 °C for 10 min
Ag (100 nm)	–	–	–	From 110 to 170 °C for 30 min

In this work, we investigated the effect of post-annealing on the performance of each layer and on the interfaces between the layers of I-PSCs by carrying out various annealing sequences prior to Ag electrode deposition: (1) after spin-coating the WPF-oxy-F layer, and annealing; (2) after spin-coating active layer on underlying WPF-oxy-F layer, and annealing; and (3) after spin-coating PEDOT:PSS on the underlying two layers, and at 170 °C for 30 min under the same post-annealing conditions (referring to Table 1). This investigation of the different annealing methods indicated that both the interfaces of P3HT:PCBM/PEDOT:PSS and PEDOT:PSS/Ag in I-PSCs with WPF-oxy-F were mainly dependent on the post-annealing.

2. Experimental details

2.1. Device fabrication and characterization

Fig. 1 shows schematics of our device structure and the chemical structure of the WPF-oxy-F. To fabricate the device, indium tin oxide (ITO) coated glass substrates (Samsung Corning Co., Ltd.) were cleaned in an ultrasonic bath sequentially with acetone, deionized water (DI) and isopropyl alcohol (IPA), and dried in an oven. A diluted solution of WPF-oxy-F in methanol was spin-coated onto the ITO substrate with a thickness of ~3 nm in a N_2-filled glove box. As an active layer, a blend solution film of 30 mg of P3HT (Rieke Metals) and 15 mg of PCBM (Nano-C) in 2 ml of chlorobenzene was then spin-coated onto the WPF-oxy-F layer to a thickness of 60 nm, followed by drying at 110 °C for 10 min in the N_2-filled glove box. PEDOT:PSS with 16 nm-thick film was spin-coated onto the active layer from IPA, followed by drying at 120 °C for 10 min in ambient air. Then, 100 nm of silver (Ag) was thermally deposited onto the PEDOT:PSS layer in a vacuum at 10^{-6} Torr. After the deposition of the Ag electrode, the devices were post-annealed at 110, 130, 150 and 170 °C for 30 min. The area of the device was 4.64 mm^2. The photocurrent–voltage (J–V) characteristics of the devices were measured using a Keithley 4200 instrument under 100 mA cm^{-2} illumination from a 1-kW Oriel solar simulator with an AM 1.5G filter in the N_2-filled glove box. A calibrated silicon reference solar cell with a KG5 filter certified by the National Renewable Energy Laboratory (NREL) was used to confirm the measurement conditions.

2.2. Thin-film characterization

The characteristics for surface roughness and morphology of the active layer and PEDOT:PSS were obtained using Atomic Force Microscopy (AFM, Digital Instruments Nanoscope IV in tapping mode). The modified Ag work-functions of PEDOT:PSS were measured using Kelvin Probe Measurements (KP 6500 Digital Kelvin probe, McAllister Technical Services. Co. Ltd). The glass substrate (Corning Eagle 2000) was cleaned in an ultrasonic bath sequentially with acetone, deionized water and IPA, and dried in an oven. Ag was thermally deposited onto the glass substrate in a vacuum at 10^{-6} Torr, and then PEDOT:PSS was spin-coated on an Ag/glass substrate, and the modified Ag was treated under the same conditions as those in the actual device fabrication. The contact potential difference (CPD) was calibrated to a highly ordered pyrolytic graphite (HOPG) at -4.58 ± 0.03 eV.

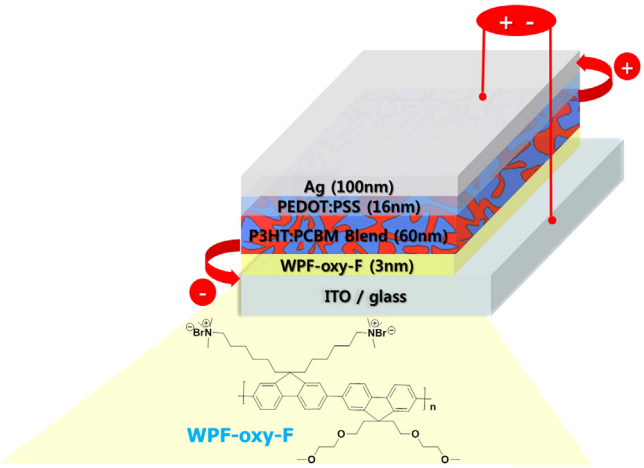

Fig. 1. Structure of the inverted polymer solar cell with a WPF-oxy-F layer.

3. Results and discussion

Fig. 2(a) shows the current density–voltage (J–V) curve of the devices treated by different treatment processes at 170 °C for 30 min. Under the post-annealing conditions, the V_{oc} and FF of the inverted device were enhanced from 0.64 to 0.68 V and from 36% to 64%, respectively, and the PCE increased from 2.13% to 3.86%. We found that post-annealing at 170 °C was the optimum annealing condition leading to high-performance devices, which were better than our Ag-based normal PSCs with configurations of ITO/PEDOT:PSS/P3HT:PCBM/WPF-oxy-F/Ag (V_{oc}=0.60 V, J_{sc}=9.17 mA cm^{-2}, FF=50% and PCE=2.77%) [20]. Although 170 °C is somewhat high for a manufacturing process, it is interesting that I-PSCs with WPF-oxy-F can endure 170 °C as opposed to I-PSCs with Cs_2CO_3 [12]. However, further increasing the temperatures (190 °C and 210 °C) leads to device performance degradation (see Fig S1) from unfavorable phase separation between P3HT and PCBM [15] or thermal decomposition of the WPF-oxy-F [38]. The photovoltaic performance is summarized in Table 2, and the device performances of each treatment process as functions of temperature are also shown in Figs. S4 and S5 and Tables S2–S4.

During the post-annealing [25,26], which means thermal annealing after deposition of all the layers, including Ag electrode, we wanted to learn which layer or interface was most effectively modified. Therefore, we performed thermal annealing sequentially after deposition of each layer, beginning with the WPF-oxy-F layer deposition, as shown in Table 1. The device performances of all devices under the **(1)** conditions (data not shown) were similar to those of devices without certain annealing treatments. Although there were no differences in the device performance, the surface energy of WPF-oxy-F of 32.4 mN/m at 170 °C, quite similar to the

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