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Pentacene-assisted planarization of photo-active layers for high performance tandem organic photovoltaics

Feng Yang[a], Dong-Won Kang[b,*], Yong-Sang Kim[a,*]

[a] School of Electronic and Electrical Engineering, Sungkyunkwan University, Suwon, Gyeonggi 16419, Republic of Korea
[b] School of Energy Systems Engineering, Chung-Ang University, Seoul 06974, Republic of Korea

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ABSTRACT

In organic tandem solar cells, the morphology of the photo-active layers is not usually stable when their fabrication processes are made with several thermal treatments. Here, we report pentacene-assisted planar tandem organic photovoltaic device based on the active layers of poly(3-hexylthiophene) (P3HT) and (6,6)-phenyl C61-butyric acid methyl ester (PCBM), fabricated in air-ambient. An additive of pentacene was introduced into the active layers of the bottom sub-cells for planarization on their surfaces. The surface morphology of pentacene-based active layer was maintained to be flat from 80 to 160 °C, and allowed for fabricating the planar active layers and interconnecting layer of tandem devices against thermal treatment. State-of-the-art homo-tandem organic solar cells were achieved with an average power conversion efficiency (PCE) of 3.5%, while their best single junction solar cells of P3HT:PCBM achieved a PCE of 3.2%. We showed the first application of high performance tandem organic photovoltaics with three times of high temperature annealing processes at 160 °C. Our work demonstrates a practical way to design highly efficient tandem organic solar cells with other powerful and well-chosen bandgap energy active layers during thermal treatments.

1. Introduction

The power conversion efficiency (PCE) of organic photovoltaics (OPVs) has been highly developed by more than 10% (Cheng et al., 2016; Ouyang et al., 2015; You et al., 2013). For higher PCE, tandem organic solar cells have attracted enormous research interest to harvest more solar energy. Recently, many papers report more than 12% PCE of tandem OPV devices (Li et al., 2017; Zhang, K. et al., 2016; Zhang, Q. et al., 2016), and the highest PCE is to date 13.8% (Cui et al., 2017). However, a theoretical maximum PCE of 40% for tandem photovoltaics is proposed by employing well-chosen bandgap energies for their active layers (Rabady and Manasreh, 2017). Thus, there is much room to improve the performance of tandem organic solar cells.

There are a lot of technologies to develop the performance of tandem OPVs, such as the synthesis of new organic materials for active layers (Dou et al., 2012; Li et al., 2016), and various designs of interconnection layer (ICL) (Ameri et al., 2013; Etxebarria et al., 2015). ICL is a very critical connector between the sub-cells of tandem devices. For an ICL, it typically consists of a hole transport layer (HTL) and an electron transport layer (ETL). Solution-processed HTLs or ETLs for ICLs are very popular approaches, such as poly(3,4-ethylenedioxythiophene): poly-styrene sulfonate (PEDOT:PSS) (Chen et al., 2014, 2013), MoO_x (Lu et al., 2015), ZnO (Kouijzer et al., 2012), or TiO_x (Kong et al., 2012; Yang et al., 2011). Some HTLs or ETLs of metal oxide nanoparticles are processed at room-temperature (Lu et al., 2015; Zhou et al., 2015). However, the layers of metal oxide nanoparticles are not robust enough for ICLs, which typically need another very thick sub-layer of ICLs to protect the underlying sub-cells from the solvents of the later processes. The thick extra sub-layer of ICLs would increase their resistance and light absorption, and decrease the PCE of tandem solar cells (Chen et al., 2015). Usually, solution-processed HTLs or ETLs prefer some thermal treatment at about 100–300 °C (Wang, K. et al., 2016). Thermally treated ICLs would be more robust and efficient for tandem devices (Yang et al., 2017a). Moreover, thermal treatment on the organic single junction solar cells significantly improved their PCE performance (Busireddy et al., 2016; Proudian et al., 2016; Wang, J.L. et al., 2016). Hence, it is highly interesting to fabricate tandem organic solar cells with thermal treatment processes.

In order to fabricate the tandem OPV devices against thermal processing, morphologically-stable bottom sub-cells under the thermal processing are very fundamental to the ICL and top sub-cells of tandem devices. Under high temperature annealing, the acceptor in the active layer of OPVs devices, typical fullerene derivative [6,6]-phenyl-C61-butyric acid methyl ester (PCBM), emerges and aggregates into

* Corresponding authors.
E-mail addresses: kangdwn@cau.ac.kr (D.-W. Kang), yongsang@skku.edu (Y.-S. Kim).

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micrometer-sized crystals on the surface of active layer (Ben Dkhil et al., 2017; Salim et al., 2016), and usually leads to defects or failure of fabrication of ICL and top sub-cell. Some studies tried to fabricate thermally stable active layers for organic single-junction solar cells, such as cross-linking of the polymer (Kim et al., 2012) or fullerene (Hsieh et al., 2010), and introducing additive molecules (Grant et al., 2017; Wang et al., 2015; Yang et al., 2015). We found that there are no papers reporting tandem organic solar cells with several thermal annealing processes at high temperature. The harsh high temperature up to 150 °C may ruin the morphology of the underlying active layer, so that Chen et al. just applied at mild post-treatment of 80 °C on the ICL of their tandem OPVs (Chen et al., 2017). In particular for a top sub-cell of tandem OPVs, the active layer was mostly fabricated with solvent annealing (Li et al., 2017) or additive treatment (Cui et al., 2017), which avoids any thermal impact on the performance of tandem devices. The performance of additive treated OPV devices strongly decreased under thermal stress (Ben Dkhil et al., 2017). In any event, it is very hard to apply them to the industrial manufacturing process, which always involves many thermal processes (Jørgensen et al., 2012). In our previous study (Yang et al., 2015), organic single-junction solar cells with the additive of pentacene were thermally stable for 24 h at 120 °C, and kept 70% of their original PCE. The surface of their active layer has few micrometer-sized crystals of PCBM after the thermal treatment. Hence, for fabricating the planar ICL and top sub-cell of tandem OPVs with multi-thermal treatment, there is a chance of a bottom sub-cell with morphological engineering of pentacene.

In this work, we developed the first application of efficient tandem organic solar cells with three times of high temperature annealing processes at 160 °C. The tandem OPV device structure and chemical structures of organic materials for active layers are shown in Fig. 1(a) and (b). The sub-cell with morphology engineering of pentacene was deeply studied from 80 to 160 °C. The planar tandem organic solar cells based on the P3HT:PCBM system achieved an average PCE of 3.54% with short-circuit current density (J_{SC}) = 4.79 mA/cm^2, open-circuit voltage (V_{OC}) = 1.18 V, and fill factor (FF) = 0.63. The PCE of tandem devices with 160 °C annealing is much comparable to the recently published homo-tandem OPVs based on the P3HT:PCBM system of 3.2–3.5% with 100 °C annealing (Lu et al., 2016, 2015).

2. Experimental section

2.1. Device fabrication

Poly(3-hexylthiophene) (P3HT) (from Rieke Metals), [6,6]-Phenyl C61 butyric acid methyl ester (PCBM) (from Nano-C) and pentacene (from Polysis Company) were used as received without further treatment. A PEDOT:PSS solution (Clevious PVP AI 4083) was modified with Triton X-100 (0.5% v/v). For single-junction cell, patterned ITO substrates were ultra-sonicated sequentially in deionized water, acetone, and then isopropyl alcohol for 20 min of each step. A ZnO layer (ca. 40 nm) was prepared on the ITO substrate (Yang et al., 2015). P3HT:PCBM:pentacene (1:1:0.2 w/w) for the active layers of various thicknesses, dissolved in chlorobenzene by ultra-sonication, was spin-coated on the ZnO layers. The modified PEDOT:PSS solution was spin-coated on the active layers, and thermally treated at 160 °C for 10 min (Yang et al., 2016). A silver (Ag) electrode (ca. 100 nm) was deposited on the PEDOT layer by a thermal evaporator. For tandem solar cells, an inter-connecting layer (ICL) of MoO_3/Au/Al/ZnO layer was deposited on the bottom sub-cells as our previous study (Yang et al., 2017a). The wet ZnO film of ICL was thermally treated at 160 °C for 10 min. The active layer, PEDOT:PSS, and Ag layer of top sub-cells were fabricated similarly as the single-junction solar cells procedure. All fabrication of single-junction cells and tandem solar cells were fabricated in the ambient air (20–40% relative humidity) except the thermal evaporation processes. The area of all cells was 0.09 cm^2, defined by the overlap area between the ITO and the Ag electrode.

2.2. Device characterization

The scanning electron microscope (SEM) images were obtained with a JEOL JSM7000F field emission scanning electron microscope (FESEM). All SEM samples were applied platinum-coating prior to SEM imaging. Atomic force microscopy (AFM) images were measured with an advanced scanning probe microscope (PSIA Corp.). The thicknesses of active layers were characterized from the average values of their AFM images. Optical microscopy images were acquired by an Olympus BX41 Microscope Digital Camera. The current density – voltage (*J–V*) characterization of solar cells was measured with a *J-V* curve tracer

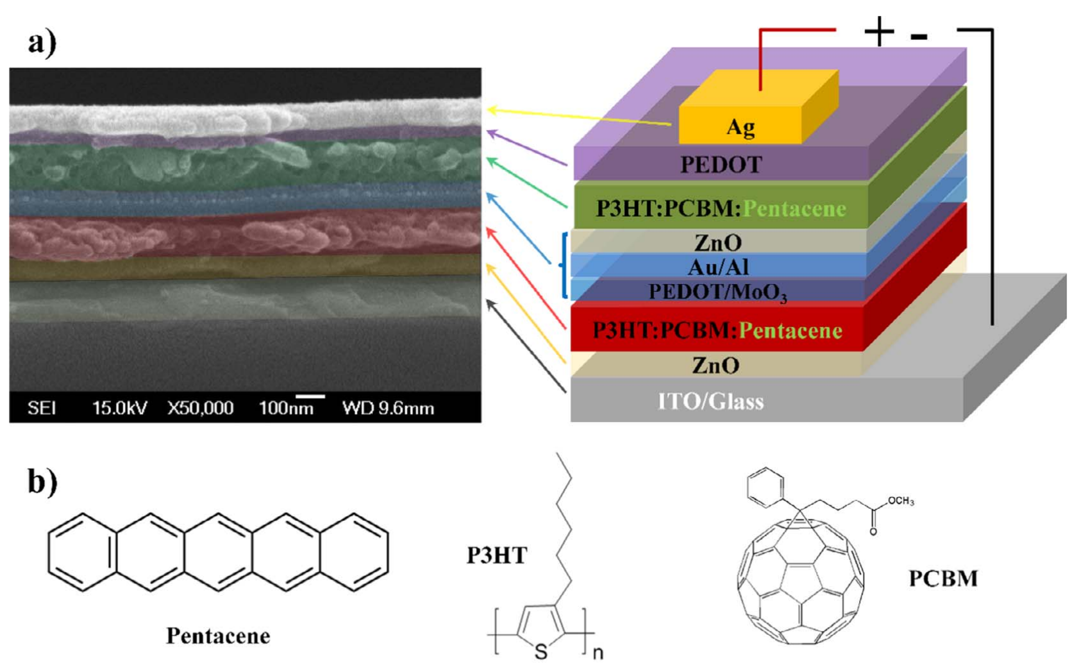

Fig. 1. (a) The cross-section SEM image (false color) and device structure of tandem OPVs: ITO/ZnO/P3HT:PCBM:Pentacene/PEDOT:PSS/MoO_3/Au/Al/ZnO/P3HT:PCBM:Pentacene/PEDOT:PSS/Ag. (b) Chemical structures of pentacene, P3HT, and PCBM in tandem OPV cells.