



An efficient and thermally stable interconnecting layer for tandem organic solar cells



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ABSTRACT

We report a novel interconnecting layer (ICL) consisting of MoO₃/Au/Al/ZnO for efficient operation of inverted homo-tandem organic solar cells employing poly(3-hexylthiophene) (P3HT) and (6,6)-phenyl C61-butyric acid methyl ester (PCBM). It was found that the ultrathin metal bilayer of Au/Al, a few nanometers, can align the energy levels and play a significant role in the charge extraction and recombination within the ICL. And the ICL showed an electrically ohmic-contact, high transmittance and very planar morphology for tandem application. The performance of air-fabricated homo-tandem organic solar cells with this new ICL delivered an average power conversion efficiency (PCE) of 3.0%, which is very close to the PCE of 3.3% from the state-of-the-art P3HT:PCBM based homo-tandem organic solar cells. Meanwhile, thermal stability tests at an elevated temperature of 150 °C for 12 h showed stable ICL with successful operation.

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1. Introduction

The photovoltaic technology is growing up to its potential as a clean and renewable energy resource. Efficient organic solar cells with the advantages of low cost, easy fabrication process, stability, and flexibility, can be the great prospects of commercial applications for organic photovoltaics (OPVs) (Ameri et al., 2013). OPVs have been widely developed with the power conversion efficiency (PCE) of more than 10% by enormous research in last five years (You et al., 2013c, 2013a; Ouyang et al., 2015). Tandem concept attracts considerable research interest to overcome the energy losses in single cell OPV devices, such as narrow absorption spectra and thermalization loss (Sista et al., 2011). It is suggested that a maximum PCE of 21% is theoretically achievable for organic tandem solar cells by combining a front cell with a bandgap energy (E_g) of ≈ 1.6 eV and a rear cell with a E_g of ≈ 1.2 eV (Li et al., 2014). With the rapid development of novel materials and engineering techniques, the PCE of tandem OPVs recently has reached about 12% (Yusoff et al., 2015; Zhou et al., 2015; Chen et al., 2014a). There is still a room for the design of the tandem cells, because of the huge difference between the theoretical maximum PCE and the recently published works. Therefore, the tandem OPV devices

should provide a promising route to realize high performance organic solar cells.

Tandem OPVs research recently has been developed on two aspects: the synthesis of new donor (Dou et al., 2012) or acceptor (Li et al., 2016) materials for more efficient light absorption, and various designs of interconnection layer (ICL) with better charge extraction and recombination (Qing et al., 2014). The design of new materials has improved a lot in single junction organic solar cells (Ameri et al., 2013; Yip and Jen, 2012; You et al., 2013b). An efficient ICL is an important issue of tandem organic solar cells. In a typical tandem device, the two single sub-cells are stacked and connected by an ICL. The ICL generally consisted of a p-type hole transporting layer (HTL) and an n-type electron transporting layer (ETL), which function as the charge extraction and recombination layers (Zhou et al., 2015). The charge recombination depends on the alignment of Fermi levels of the HTL and ETL. There are some principles for the design of ICL. (1) ICL must have a high optical transmittance to minimize optical losses; (2) between the ETL and HTL, a quasi-ohmic-contact should be made to allow for electron-hole charge extraction and recombination; (3) the ICL should be robust enough to protect the underlying active layer from dissolution by the processing solvents for the top cell; and (4) The ICL should be environmentally stable to enhance the stability and lifetime of the tandem device (Ameri et al., 2013).

Many ICLs of tandem organic solar cells are reported. The typical ETLs in tandem OPV devices include transparent transition

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metal oxides, such as ZnO (Chen et al., 2013; Kouijzer et al., 2012), TiO₂ (Yang et al., 2011; Kong et al., 2012) and polymer dipole layers (Yusoff et al., 2014; Zhou et al., 2012; Shim et al., 2012; Lu et al., 2015), such as PFN, CPE, PEIE. For HTL, metal oxides such as V₂O₅ (Chou et al., 2011), MoO₃, (Fan et al., 2013; Liu et al., 2014), and conductive polymers, such as PEDOT:PSS (Chang et al., 2012), are widely adopted in tandem organic solar cells. Also, some groups reported combined evaporation and solution approach ICLs such as MoO₃/Al/ZnO (Chou et al., 2011), and GO-Cs/Al/GO/MoO₃ (Chen et al., 2014b). Moreover, there are examples of evaporation based ICLs such as Ca/Ag/MoO₃ (Fan et al., 2013), MoO₃/Ag/Al/Ca (Sun et al., 2010), and BCP/Ag/ReO₃ (Shim et al., 2014). However, efficient and thermally stable ICL is still a challenge for tandem organic solar cells. There are no papers studying on the thermal stability of ICL, but one paper (Kong et al., 2012) reported that they conducted the post-annealing with less than 150 °C for a few minutes. The post-annealing process is thought to induce an electrokinetic migration in which the mobile positive ions move toward/over the interface between TiO_x and PEDOT:PSS. As a result, a dipole layer could be formed in between the n-/p-type interlayers, originating from electrostatic acid-base interaction. It seems that the thermal stability of dipole layer in the ICL is sensitive or not much studied. To date, our research for first time reported the thermal stability of organic tandem solar cells.

In this work, we propose a new ICL of PEDOT:PSS/MoO₃/Au/Al/ZnO for tandem OPV devices, which is an efficient and thermally stable combination of different HTLs, metals, and ETL. For MoO₃ layer of the ICL, its performance is highly related to the oxygen vacancies which is not much stable when exposed in the air. Energy level alignment can be achieved in MoO₃/Au through Fermi level pinning transition. The Au/Al bilayer effectively aligns the energy levels in the ICL. With metal-metal contact, the Fermi levels of Au and Al were aligned without any energy barrier and were beneficial for thermal resistance in this complex ICL structure of PEDOT:PSS/MoO₃/Au/Al/ZnO. Here, an ultrathin metal bilayer of Au/Al was suggested as a function of the charge recombination by aligning the energy levels and maintaining a high transmittance. The inverted tandem organic solar cells based on poly(3-hexylthiophene) (P3HT) and (6,6)-phenyl C61-butyric acid methyl ester (PCBM) were fabricated by sandwiching the suggested ICL. It should be noted that the tandem cells were prepared without glovebox using inert gases since the glovebox-based process is expensive and laborious compared to the process under air-ambient (Sun et al., 2011). The performance of the air-fabricated homo-tandem organic solar cells has an average PCE of 3.0%, which is very close to the PCE of 3.3% from the state-of-the-art P3HT:PCBM based homo-tandem organic solar cells (Lu et al., 2015). And the test of thermal stability at 150 °C for 12 h showed stable ICL with successful operation.

2. Experimental section

2.1. Materials

The indium tin oxide (ITO) coated glass was supplied by Fine Chemicals (South Korea) (15 Ω/square sheet resistance). Zinc acetate dihydrate (99.999%), monoethanolamine (MEA, ACS reagent, 99.0%), and 2-methoxyethanol (2-ME, anhydrous, 99.8%) were from Sigma Aldrich. Poly(3-hexylthiophene) (P3HT) was purchased from Rieke Metals and [6,6]-Phenyl C61 butyric acid methyl ester (PCBM) from Nano-C. Chlorobenzene, anhydrous, 99.9% was from Sigma Aldrich. PEDOT:PSS (Clevios PVP Al 4083) was supplied by Hoescht Deutschland GmbH & Co. KG (Germany). Triton X-100, extra pure, was supplied by Do Chemical Co., Ltd. Hexamethyldisilazane (HMDS) was purchased from AZ Electronic Materi-

als (South Korea). Gold (Au) was from Vacuum Thin Film Materials Co. (South Korea) and Aluminum (Al), silver (Ag), MoO₃ (99.999%) from Alfa Aesar.

2.2. Single cell fabrication

The patterned ITO substrates were ultra-sonicated for 10 min in deionized water followed by acetone and then isopropyl alcohol. A 40 nm ZnO film, spin-coated from a ZnO precursor solution, was obtained on the ITO substrate (Yang et al., 2015). A 1:0.7 w/w blend of P3HT and PCBM, with the concentration of 10 mg/ml for P3HT, was dissolved in chlorobenzene by ultra-sonication for 4 h, filtered through a 0.45 μm PVDF filter, and spin-coated on the ZnO-coated ITO substrates. The formed active layer of P3HT:PCBM was ~60 nm in thickness. PEDOT:PSS solution was modified with 0.5% v/v of Triton X-100 nonionic surfactant (Yang et al., 2016). HMDS was first spin-coated on the active layer, followed by the PEDOT:PSS deposition. The modified PEDOT:PSS was spin-coated on the active layer and annealed in an oven at 160 °C for 10 min. A PEDOT:PSS film of 30–40 nm was obtained on the active layer. The devices were prepared for measurement after the thermal deposition of ~100 nm thick Ag film through a shadow mask. All fabrication processes were carried out in the ambient air except vacuum thermal deposition process of silver. The active area of each device was defined by the overlap between the patterned ITO and Ag electrodes, 0.09 cm².

2.3. Tandem device fabrication

The front cells were made according to the single cells procedure before deposition of Ag, and then the samples were transferred into the evaporation chamber. 10 nm MoO₃, 3 nm Au and 2 nm Al (without breaking the vacuum) were subsequently thermally evaporated on top of the front cell at 6 × 10⁻⁶ Torr. An amorphous ZnO layer was deposited with a precursor solution consisting of 0.3 M zinc acetate and 0.3 M monoethanolamine in 2-methoxyethanol at a spin speed of 4000 rpm for 40 s. The film was annealed at 150 °C for 10 min in ambient air. The active layer and PEDOT layer of top sub-cells were made according to the single cells procedure. Finally, 100 nm of silver as the anode was thermally evaporated through a shadow mask. The device area, as defined by the overlap between the patterned ITO and Ag electrodes, was 0.09 cm². 1:1:0.1 (or 1:1:0.2) w/w blends of P3HT:PCBM:pentacene for active layer of modified the sub-cells were dissolved in chlorobenzene by ultra-sonication of 4 h, filtered with a 0.45 μm PVDF filter, and spin-coated on the ZnO film.

2.4. Device characterization

The current density - voltage (*J-V*) characterization of devices was measured with *J-V* curve tracer (Eko MP-160) and a solar simulator (Yss-E40, Yamashita Denso) under AM 1.5G irradiation with the intensity of 100 mW/cm², calibrated by Newport certified standard silicon cell. The transmittance spectroscopy measurements were made over a wavelength range of 200–900 nm using a Shimadzu UV-1601 UV-Vis spectrophotometer (the transmittance of 0.7 mm thick of glass substrate was measured as the base). Optical microscopy images were obtained using an Olympus BX41 Microscope Digital Camera. Atomic force microscopy (AFM) images were obtained with an advanced scanning probe microscope (PSIA Corp). The thicknesses of all films were characterized by the AFM system. The thermal stability measurements of tandem solar cells were taken at an elevated temperature of 150 °C on a hotplate in ambient air.

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