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Large active area inverted tandem polymer solar cell with high performance via insertion of subnano-scale silver layer

Po-Nan Yeh [a,b], Tzu-Hao Jen [a,b], Yu-Shan Cheng [a,b], Show-An Chen [a,b,*]

[a] Chemical Engineering Department, National Tsing-Hua University, Hsinchu 30013, Taiwan, Republic of China
[b] Frontier Research Center on Fundamental and Applied Sciences of Matters, National Tsing-Hua University, Hsinchu 30013, Taiwan, Republic of China

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ABSTRACT

We propose the deposition of ultra-thin silver layer (in subnano-scale) between bottom subcell and the hole-transport sublayer in the interlayer (IL), poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS), in the inverted tandem polymer solar cell (t-PSC) for improving the wettability of aqueous hole-transport interlayer on hydrophobic active layer so that the performance is significantly improved and larger active area is allowed. The inverted t-PSC is composed of bottom subcell (regioregular poly (3-hexylthiophene) (P3HT): indene-C_{60}-bisadduct (ICBA)) and the top subcell (thieno[3,4-b]thiophene/benzodithiophene (PTB7) with [6,6]-phenyl C_{71}-butyric acid methyl ester ($PC_{71}BM$)) along with the IL, PEDOT:PSS/ZnO nanoparticles. For the small-area (0.03 cm^2) t-PSC, the power conversion efficiency (PCE) can be improved from 7.06% without Ag layer to 7.81% with depositing 0.5 nm Ag layer, and the device production yield from 40% (4 out of 10 devices) to near 100% (10 out of 10 devices), which can be ascribed to more efficient hole collection and reducing leakage current for the bottom subcell due to improvement of film forming of PEDOT:PSS. As the active area is increased to 1 cm^2, a PCE of over 6% (6.11%) can also be achieved by this approach, which is much higher than that without such Ag layer (2.19%) by a factor of 2.78. The reported active area of inverted t-PSC is in the range 0.02–0.15 cm^2; while the present method allows an inverted t-PSC to enlarge its active area to 1 cm^2, yet the PCE can maintain over 6%.

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1. Introduction

The development of new energy sources is of growing importance due to the global energy crisis. The utilization of conjugated polymer as an active layer of solar cell has attracted great attention in recent years due to the ease of fabrication, promising flexibility and low cost production. The bulk heterojunction polymer solar cell (BHJ-PSC) consisting of regioregular poly(3-hexylthiophene) (P3HT) as a donor and [6,6]-phenyl-C_{61}-butyric acid methyl ester (PCBM) as an acceptor is the most intensely investigated system, giving the power conversion efficiencies (PCEs) of 4–5% [1]. To harvest more sunlight as compared to P3HT, various low-bandgap conjugated polymers have been proposed [2–5]. Recently, the PCE of BHJ-PSC based on the composite film of low-bandgap conjugated polymer thieno [3,4-b]thiophene/benzodithiophene (PTB7) with [6,6]-phenyl C_{71}-butyric acid methyl ester ($PC_{71}BM$) can reach the PCE 9.2% [6].

Although the utilization of low-bandgap conjugated polymer can improve absorption range of solar spectrum, it suffers from inherent disadvantage of low open-circuit voltage (V_{OC}) (0.65–0.75 V) determined by the highest occupied molecular orbital (HOMO) level of polymer [7–9], limiting its practical application. A promising solution for this problem is to employ the tandem cell architecture. The tandem PSC (t-PSC) consists of two or more subcells (each subcell is a BHJ-PSC) having complementary absorption in series connection via an interlayer (IL). Apart from covering a broader absorption range of the solar spectrum, the V_{OC} of t-PSC is close to the summation of the V_{OC}'s of the individual subcells. Therefore, high V_{OC} can be expected in the t-PSC. So far, PCE of 8.6% with V_{OC} of 1.5 V can be achieved in a conventional t-PSC [10], and those for inverted t-PSC are 10.6% and 1.53 V [11], respectively, where indium–tin oxide (ITO) acts as the cathode rather than the anode in the inverted t-PSC. Form a practical point of view, the inverted device structure is an ideal architecture for BHJ-PSC. In contrast to the utilization of air-sensitive low-work-function metal in the conventional device, the air-stable high work function metal used in the inverted device can improve its long-term stability. As a result, combining tandem and inverted structure in PSC will benefit in terms of range of sunlight harvesting, device efficiency, and device stability.

In all of the reported IL for the inverted t-PSC [10–13], the IL composed of poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) and ZnO nanoparticles (NP) layers is the most

* Corresponding author at: Chemical Engineering Department, National Tsing-Hua University, Hsinchu 30013, Taiwan, Republic of China. Tel.: +886 3 571 0733; fax: +886 3 573 7798.
E-mail address: sachen@che.nthu.edu.tw (S.-A. Chen).

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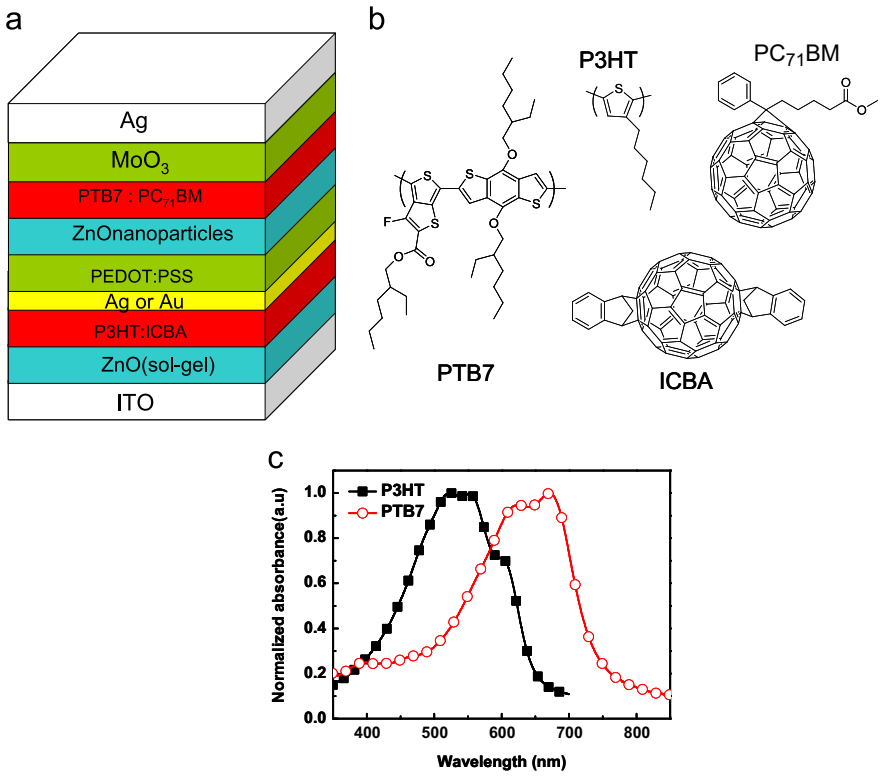

Fig. 1. (a) Device structure of inverted t-PSC, (b) chemical structures of PTB7, P3HT, ICBA and $PC_{71}BM$ used in the active layers and (c) normalized UV–vis spectra of the thin films of P3HT and PTB7.

attractive one due to their solution-processing capability. However, the aqueous solution of PEDOT:PSS does not wet the surface of active layer very well due to hydrophobic nature of the active layer [14], Therefore, the active areas of the reported inverted t-PSCs based on PEDOT:PSS/ZnO NP IL are usually very small, ranging from 0.02 to 0.15 cm^2 [10–14], which will lead to serious limitation for practical application. Although it has been reported that N_2 plasma treatment of active layer surface can improve film formation of PEDOT:PSS [14], the use of N_2 plasma during device fabrication can inevitably damage the active layer.

In this contribution, for the first time, we demonstrate the deposition of ultra-thin metal layer (in subnano-scale) of Ag between bottom subcell and PEDOT:PSS in the inverted t-PSC to improve wettability of PEDOT:PSS aqueous solution on hydrophobic active layer. The device structure of the inverted t-PSC is ITO/ZnO/P3HT:indene-C_{60}-bisadduct (ICBA) (1:1, w/w)/Ag (x nm)/PEDOT:PSS/ZnO NP/PTB7:$PC_{71}BM$ (1:1.5, w/w)/MoO_3/Ag (its schematic diagram and chemical structures of the materials used in the active layers are shown in Fig. 1). The selection of these two conjugated polymers (P3HT and PTB7) is due to their complementary optical absorptions of the solar spectrum (Fig. 1).

2. Material and methods

2.1. Materials and general instrumentation

Regioregular poly(3-hexylthiophene) (P3HT, 4002-Egrade, Rieke Metals, regioregularity 97.8%), thieno [3,4-b]thiophene/benzodithiophene (PTB7, One Material), indene-C_{60}-bisadduct (ICBA, Nano-C), [6,6]-phenyl C71 butyric acid methyl ester ($PC_{71}BM$, 99.5%, Nano-C) and 1,8-diodooctane (98.0%, Sigma-Aldrich) were used as received. Ultraviolet–visible (UV–vis) spectra were measured using an UV–vis spectrometer (Hitachi U3300). The thickness of polymer film was measured by a Tencor P-10 Surface Profiler. A contact angle meter employing high resolution cameras was used to capture and analyze the contact angle between the PEDOT:PSS and active layer interface.

2.2. Atomic force microscopy (AFM) measurement

The AFM (Digital Instrument Nanoscope V) was performed under ambient atmosphere. Rectangular Au tips (Multi75GB, Budget Sensors) with an estimated force constant of 3 N m^{-1} and resonance frequency of 75 kHz were used in tapping mode for height and phase images.

2.3. Device fabrication and photocurrent measurement

The fabrication procedure for the inverted tandem polymer solar cells (t-PSC) is as follows. The ITO glass plates were cleaned sequentially with acetone, isopropyl alcohol, mixed solvent of deionized water/H_2O_2/NH_4OH (5:1:1, v/v/v), deionized water, and acetone. Thin film of ZnO was prepared similar to the previous report [15], in which ZnO precursor solution was spin-coated at 5000 rpm on top of the cleared ITO and then heating at 200 °C for 1 h in air, giving transparent ZnO thin film with the thickness about 30 nm. The ZnO precursor solution was prepared by dissolving zinc acetate dehydrate $C_4H_6O_4Zn{\bullet}2(H_2O)$ (99.5%, Merck, 1 g) and monoethanolamine ($HOCH_2CH_2NH_2$, 98%, Acros, 0.28 g) in 2-methoxyethanol ($CH_3OCH_2CH_2OH$, Aldrich, 99.8%, 10 mL) under stirring for 8 h for hydrolysis reaction and aging. On top of the ZnO layer, a thin layer (about 180 nm) of P3HT:ICBA (1:1, w/w) was deposited by spin-casting from its solution (1.7 wt %) in 1,2-dichlorobenzene in argon-filled glove box and dried in covered glass petri dishes for 2 h. After thermal annealing at 150 °C for 10 min, the sample was then moved to atmosphere for subsequent spin-coating of 85 nm-thick PEDOT:PSS (Clevios P VP AI4083), in which the surfactant of triton X-100 (1 wt%) (laboratory grade, Sigma-Aldrich) was added to the aqueous solution of PEDOT:PSS to improve wetting of PEDOT:PSS on the P3HT:ICBA surface. (The weight ratio of solid content of PEDOT:PSS aqueous solution to triton X-100 is

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