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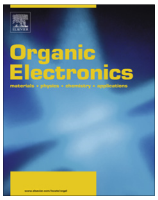

Performance enhancement in inverted polymer solar cells incorporating ultrathin Au and LiF modified ZnO electron transporting interlayer

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ABSTRACT

The performance enhancement of inverted polymer solar cells (PSCs), based on the blend system of regioregular poly(3-hexylthiophene) and [6,6]-phenyl C_{61}-butyric acid methylester, due to incorporating ultrathin Au and LiF interlayer between the front transparent indium tin oxide and a ZnO electron transporting layer was analyzed. The results reveal that a 40% increase in PCE, e.g., from 2.62% to 3.67%, was observed for PSCs made with an optimal Au/LiF interlayer as compared to the one having a bare ZnO electron transporting layer. The presence of Au/LiF-modified ZnO interlayer between ITO and the organic layer helps to improve the charge collection. The absorption enhancement arising from the plasmon resonance of Au nanostructures also contributed to the improvement in PCE. It is shown that PSCs with LiF incorporated ZnO electron transporting layer allow improving cell lifetime, demonstrating <50% decrease in PCE compared to that of the ones with a bare ZnO interlayer after 240 day aging test for cells without encapsulation in air.

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1. Introduction

Considerable effort has been devoted to the development of polymer solar cells (PSCs) as a promising alternative photovoltaic technology. Bulk heterojunction PSCs based on low band-gap conjugated polymer and phenyl-C_{70}-butyric acid methylester with power conversion efficiency (PCE) of ~10% were reported [1,2]. However, the PCE of PSCs is still limited by its relatively low carrier mobility. The absorption in PSCs is limited due to a mismatch between light absorption depth and carrier transport scale [3]. Several light trapping techniques, such as diffraction grating, tandem structures and incorporating metal nanostructures, have been developed to increase the optical absorption of solar cells [4].

The absorption enhancement in PSCs incorporating metal nanostructures has been demonstrated due to its enhanced near-field and forward light scattering caused by the nanostructures [4–6]. In previous reports, metal nanostructures are usually incorporated into the buffer and photoactive layer, inserted between tandem cells, and attached onto the electrodes [4,5,7–10]. The plasmonic enhancement of PSCs is partially contributed by the incorporation of metal nanostructures [7,10,11]. Meanwhile, our reports have shown the deteriorating performance of

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PSCs made with metal nanostructures due to a higher serial resistance and lower shunt resistance. This increases interfacial barrier at organic/electrode interfaces, metal nanoparticle-induced carrier traps and carrier combinations [12]. The improved electrical conductivity of buffer layer and photoactive layer is considered as an important factor for enhancing PCE in PSCs [8,9,13]. Kim et al. reported 50–70% improvement in PCE of PSCs, which is primarily attributed to the improved electrical conductivity of the photoactive layer doped with Au and Ag nanoparticles [8]. Theoretical simulations showed that the incorporation of metal nanostructures in PSCs can greatly increase the optical absorption of photoactive layers [5,6]. Optimal carrier collecting/transporting layers are used to inhibit defects in photoactive layers penetrated into the whole PSCs [14], which are very important for the application of PSCs with nanoparticles. Solution-processed ZnO nanoparticles are commonly used as a good electron transport/collecting layer in inverted PSCs [15,16] due to their relatively high transparency, high electron mobility and air stability. Recently, fullerene derivative [17], conjugated polyelectrolytes [18], poly(ethylene oxide) [19], ionic liquid functionalized carbon nanoparticles [20,21] and cesium stearate [22] have been reported to modify ZnO layer to optimize contact properties between ZnO, photoactive layer and passivate traps of bulk ZnO layer, improving the photocurrent and fill factor of PSCs. In a related work, we found that the dissociation of excitons at the electrode/organic interface in PSCs plays an important role in determining the charge collection in regular and inverted PSCs. The unfavorable exciton dissociation at the organic/electrode interface for charge collection can be eliminated by interposing a thin metal oxide interlayer between the organic layer and electrode [23]. In this work, the presence of a dual interlayer consisting of ultrathin Au and LiF layers between ITO and ZnO is able to improve the contact quality of ZnO layer for application in inverted PSCs. The enhancement in PCE of PSCs with ZnO layer, modified with Au/LiF and LiF layers, is observed compared to that of control PSCs made with bare ZnO layer. In addition to the absorption enhancement due to plasmonic effect, the improvement in the contact quality at ZnO/ITO interface also results in cells with a lower serial resistance, which is contributed by the Au/LiF bilayer leading to a significant enhancement in the performance of PSCs.

2. Experimental

Bulk heterojunction PSCs, based on poly(3-hexylthiophene) (P3HT) and [6,6]-phenyl C_{61} butyric acid methyl ester (PCBM) blend system, were fabricated. The pre-patterned ITO/glass substrates, having a sheet resistance of 10 Ω/□, Q5 were cleaned by ultrasonication sequentially with detergent, de-ionized water, acetone and isopropanol for 15 min. After being dried in the oven, ITO glass substrates were treated with ultraviolet ozone for 15 min before spin-coated with a layer of ZnO, Au and LiF layers were thermally deposited on ZnO under the base pressure of 6.0×10^{-4} Pa. A ~20 nm thick ZnO electron transport/collecting layer was spin-coated on top of ITO/glass or Au/LiF-modified ITO/glass substrates, followed by annealing at 140 °C for 10 min in air. The ZnO nanoparticles were prepared as described in the previous paper [24,25]. The typical synthesis, a stoichiometric amount of tetramethylammonium hydroxide dissolved in ethanol (0.5 M) was gradually dropped into 0.1 M zinc acetate dihydrate dissolved in dimethyl sulfoxide (DMSO), followed by stirring for an hour at room temperature. After being washed with hexane and ethanol (2:1) mixing solvents, ZnO nanoparticles were dispersed in ethanol. Solutions containing P3HT:PCBM (10 mg/ml:8 mg/ml) dissolved in 1,2-dichlorobenzene were spin-coated on top of a layer of ZnO to form a 60 nm ultrathin photoactive layer in Ar-purged glove box, and then kept in a petri dish for slow growth. After 2 h, P3HT:PCBM films were annealed at 120 °C for 10 min. Finally, a 10 nm thick MoO_3 and 100 nm Al electrode were thermally evaporated onto P3HT:PCBM films in the chamber with the base pressure of 6.0×10^{-4} Pa to form an anode. The active area of the devices is 0.10 cm^2. Current density–voltage (J–V) characteristics were measured under AM1.5G illumination intensity of 100 mW/cm^2 using a Newport solar simulator system. All samples were measured in air without encapsulation. The work function of modified ITO films was measured using ultraviolet photoelectron spectroscopy (UPS) with He-I line (21.2 eV).

3. Results and discussion

The thin layer of Au, deposited by thermal evaporation on ITO surface, is easily aggregated forming Au islands due to the dewetting effect, and further formed into larger islands after annealing [26,27]. SEM images measured for Au films and ZnO films on ITO surface are shown in Fig. 1(a)–(d). It is clear that Au nanoparticles formed after a 0.9 nm thick Au layer was annealed at 140 °C. The size of Au nanoparticles onto ITO coated glass is 10–15 nm. Although the Au granules, overlapped with ZnO, cannot be observed directly in the SEM images, the size of Au granules formed on the ITO surface is expected to be larger than 15 nm due to the Au metal agglomerations which can be partially blocked by the ZnO films in the SEM measurements [27]. The contrast of SEM images measured for Au/LiF modified ZnO films increases substantially as compared to that obtained for LiF/ZnO films. The lighter areas mean higher electrical conductivity [7], indicating the existence of Au islands under ZnO films. The presence of the ZnO layer also helps to avoid the exciton recombination on the Au islands from the photoactive layer [13].

The absorption spectra of ZnO and Au/LiF modified ZnO films are shown in Fig. 1(e). The absorption spectrum of the LiF-modified ZnO film is similar to that of the ZnO film, indicating that a thin LiF film cannot affect the optical properties of ZnO electron transport layers. The absorption of Au/LiF modified ZnO films increases significantly as compared to the bare ZnO films. The protruding absorption at 530 nm of Au/LiF modified ZnO film indicates that the evaporated Au film overlapped with ZnO film were further agglomerated larger islands after annealing 140 °C for 10 min. Fig. 1(e) shows the absorption spectra of P3HT:PCBM formed on different ZnO layers. The absorption

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