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Low-temperature solution-processed ionic liquid modified SnO₂ as an excellent electron transport layer for inverted organic solar cells

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ABSTRACT

SnO₂ with its excellent properties such as high optical transparency, suitable band energy and high electron mobility, recently has received special attention from researchers as an outstanding electron transport layer (ETL) for optoelectronic devices. Here, we demonstrate that SnO₂ combined with ionic liquid (IL), 1-benzyl-3-methylimidazolium chloride ([BzMIM]Cl), based on low-temperature solution-processed can be an excellent ETL for efficient inverted organic solar cells (iOSCs). Our best performance with P3HT:PC₆₀BM based iOSCs using SnO₂/IL as an ETL, has achieved a power conversion efficiency (PCE) of 4.05%, which is the highest reported value so far and was a 38% increase compared to that of SnO₂ only (2.94%). Using ultraviolet photoelectron spectroscopy, we found that the work function of the cathode decreased significantly from – 4.38 eV to – 3.82 eV for SnO₂/IL, an outstanding feature that is necessary for an ideal ETL. Electrochemical impedance spectroscopy studies revealed a significant lower transport resistance and an efficient charge extraction at the interface between photoactive layer and the electrode for the SnO₂/IL-based iOSC than for the SnO₂ only device. The iOSC devices using SnO₂/IL showed excellent long-term stability, with a PCE of ~ 81% compared to the initial value after storage for 2.5 months in ambient conditions. This low-temperature solution-processed SnO₂/IL is expected for low-cost, high throughput, roll-to-roll process on flexible substrates for iOSC as well as other optoelectronic devices.

1. Introduction

Organic solar cells (OSCs) are third-generation photovoltaic technology that offer unique advantages, including fabrication from abundant material resources, light-weight, large-scale applicability, high throughput production at a low cost, and low-temperature solution processability that allows for fabrication on flexible substrates via roll-to-roll manufacturing [1,2]. In the last few years, OSCs have made significant progress, with their power conversion efficiency (PCE) exceeding 12% for both single junction and tandem OSCs along with a great improvement in device stability [3–5].

In general, conventional OSCs are fabricated on ITO-glass substrates deposited with PEDOT:PSS [poly(3,4-ethylenedioxythiophene):poly(styrene sulfonate)] or metal oxides as hole collection layers which function as the transparent anode, and a low work function (WF) metal, such as Ca or Al, is thermally evaporated on the back side as the cathode [6,7]. Unfortunately, the PEDOT:PSS layer exhibits acidic and hygroscopic properties, particularly at elevated temperature and

humidity, leading to corrosion in the ITO anode [8]. In addition, the low WF cathode is also sensitive with oxygen and moisture in ambient air. Therefore, both lead to a low device efficiency and poor device stability of the OSCs [9]. To circumvent such problems, an inverted device structure has been developed with a reverse charge collection [10]. In inverted organic solar cells (iOSCs), ITO or FTO have been used for the cathodes while a high WF, air-stable metal [such as copper (Cu), gold (Au), or silver (Ag)] has been used as the anode [11,12]. Therefore, iOSCs can avoid the utilization of PEDOT:PSS anode buffer layer and the low WF metal cathode, making it possible to use solution-based technology, such as printing, instead of conventional high-cost thermal evaporation that is normally used to deposit metal cathodes. At this point, iOSCs possess massive commercial potential when compared to conventional OSCs [12].

To facilitate electron transport and collection for iOSCs, various materials have been introduced and incorporated between the cathode and photoactive layer, and this has been named the electron transport layer (ETL) [13]. It is important to note that the ETL plays an important

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role in the device performance due to its transparency, and its energy level alignment can directly affect the electron extraction process. The ETL not only helps enhance the PCE, but it also improves the stability of the iOSCs, thus the ETLs are of the utmost importance [13,14]. Among various materials and technologies that are used to deposit ETLs [12], low-temperature solution-processed metal oxides have been recognized to be sufficiently reliable for commercial iOSCs due to their ease of fabrication processing, excellent stability, high transparency, and good charge transport [15–17]. So far, TiO_x and ZnO have been extensively used as ETLs [18,19], and their iOSCs using P3HT:PCBM as a photoactive layer usually yield a PCE of around 3.5% [20,21]. Unfortunately, iOSCs using these metal oxides are susceptible to “light-soaking”, i.e., UV exposure is necessary for iOSCs to work properly. Therefore, this may prevent them from being used as ETLs in the future [22,23]. Without UV treatment, these iOSCs have a tendency to exhibit S-shaped J - V curves, causing an extremely low fill factor (FF) and a poor PCE [22,23]. At this point, new metal oxide-based ETLs free of light soaking issues are needed for iOSCs.

SnO_2 is an abundant, transparent semiconducting material with a wide-band gap (3.6 eV), excellent stability, and high conductivity. Recently, SnO_2 has received a special attention from researchers as ideal ETLs for optoelectronic devices [24,25]. iOSCs using SnO_2 as ETL have been reported to be free from light soaking issues while offering excellent device stability when compared to TiO_x , ZnO, and others metal oxides [26–30]. As such, SnO_2 possesses huge potential as a choice of ETL for use in commercial iOSCs. However, the PCE of P3HT:PC₆₀BM-based iOSCs using SnO_2 or modified SnO_2 as ETLs has been reported to be around 3%, regardless of deposition techniques such as vacuum or solution process [26–30]. Thus, studies to improve the PCE of iOSCs based on SnO_2 are of utmost importance.

Researchers have used several methods to modify the ETL to improve the PCE, eliminate the light soaking issue, and improve the device stability for iOSCs, including the use of doping [31], a bilayer structure [32], inorganic-organic complexes [33], or alkali metal carbonate [34], alkali hydroxide [35], or barium hydroxide modifications [36]. Among these, ionic liquid (IL) is considered as the best choice because of its excellent physico-chemical properties like good thermal stability, high conductivity, and high electron mobility [37]. IL is known as a green chemistry that is useful in batteries, actuators, supercapacitors, and hybrid organic-inorganic optoelectronic devices [38,39]. IL has been reported to be modified or independently function as ETLs for hybrid optoelectronic devices, especially in polymer solar cells to improve their performance [40–42].

In this study, we demonstrate that IL, 1-benzyl-3-methylimidazolium chloride ([BzMIM]Cl), combined with SnO_2 can be fabricated with a low-temperature solution-process and is an excellent ETL for iOSCs. The P3HT:PC₆₀BM-based iOSCs fabricated with SnO_2 /IL showed a significantly higher average PCE of 3.94%, which is the highest reported value so far, compared to that of SnO_2 only (2.78%). By forming spontaneous dipole polarization at the interface between the cathode and the photoactive layer via SnO_2 /IL, the WF of the cathode decreased significantly from -4.38 eV (ITO/ SnO_2) to -3.82 eV (ITO/ SnO_2 /IL). As a result, the electrical contact between the ETL and the photoactive layer shifted from the Schottky contact to the Ohmic contact to contribute to an improvement in the electron transport properties of the ETL. Compared to SnO_2 only device, the enhancements in device performance of the iOSCs using IL mainly contributed to an improvement in the current density (J_{sc}) and FF. We ascribe these results to a reduction in the work-function, a better contact with a photoactive layer, and improved electron transport properties of SnO_2 /IL in iOSCs. Electrochemical impedance spectroscopy studies revealed a significant lower transport resistance and an efficient charge extraction at the interface between photoactive layer and the electrode for the SnO_2 /IL-based iOSC than for the SnO_2 only device. Furthermore, iOSCs using SnO_2 /IL showed excellent long-term stability with a remaining PCE of 81% compared to its initial value after 2.5 months of storage in ambient

conditions. This low-temperature solution-processed SnO_2 /IL may be applied in low-cost, high throughput manufacturing with a roll-to-roll process using flexible substrates to fabricate optoelectronic devices.

2. Experimental section

2.1. Materials

Absolute ethanol [$\text{C}_2\text{H}_5\text{OH}$, 99.9%] was purchased from EMD Millipore Corporation, tin (II) chloride dihydrate [$\text{SnCl}_2 \cdot 2\text{H}_2\text{O}$, 98%], and 1-Benzyl-3-methylimidazolium chloride ([BzMIM]Cl, 97%) were purchased from Alfa Aesar, and methanol [CH_3OH , 99.8%] was obtained from Sigma-Aldrich. These chemicals were used as received and without purification to prepare the SnO_2 and IL solution to fabricate the ETLs.

The donor polymer: poly(3-hexylthiophene-2,5-diyl) (P3HT) was received from Rieke Metals, Inc., and the acceptor polymer, phenyl-C₆₀-butyric acid methyl ester (PCB₆₀M) was obtained from Nano Holding. All chemicals were used as received and without purification.

2.2. Preparation and characterization of SnO_2 and SnO_2 /IL ETLs

The SnO_2 was first prepared by dissolving $\text{SnCl}_2 \cdot 2\text{H}_2\text{O}$ in absolute ethanol to a concentration of 0.1 M, followed by stirring in an oil bath at 80 °C for 8 h. To this end, after being cooling, the solution was taken away from the oil bath and kept at room temperature without further stirring. Afterward, this solution was used to prepare SnO_2 ETL without purification. IL solutions with a concentration of 0.10, 0.15, 0.20, and 0.30 wt% were prepared by dissolving [BzMIM]Cl in methanol and were then stirred at room temperature for 3 h prior to spin-coating. The patterned ITO-coated (~ 125 nm) glass substrates with a sheet resistance of ~ 12 Ω /square and a size of 2.5×2.5 cm² obtained from Korea Electronics Technology Institute were first cleaned in a sequence using an ultrasonic bath with DIO (deionized water), followed by acetone, and with isopropanol for 20 min each. Before spin-coating ETL, the ITO-glass substrates were dried using a heat gun. SnO_2 ETL with a thickness of ~ 20 nm was firstly prepared by spin-coating SnO_2 precursor solution on the ITO-glass substrates at a spin-coating speed of 3000 rpm for 40 s. The samples were annealed at 180 °C for 60 min in ambient air and were then moved into an N_2 -filled glovebox to coat the IL. For SnO_2 /IL ETL, the IL was prepared by spin-coating its solution on top of SnO_2 at a spin-coating speed of 4000 rpm for 40 s, then drying at 100 °C for 10 min on a hot plate.

The UV-vis absorption and transmission spectra of SnO_2 and SnO_2 /IL ETLs were conducted using a Shimadzu UV-2550 spectrophotometer at room temperature. Structural properties of SnO_2 and SnO_2 /IL were determined via X-ray diffraction (XRD) measurements (30 kV, 20 mA, $\text{Cu K}\alpha$ radiation with a wavelength $\lambda = 1.5406$ Å, with a scan range of 5° to 80°, a step size of 0.0334 and a time per step was 70.485). The surface composition and chemical states of the SnO_2 and SnO_2 /IL ETLs were recorded by XPS (X-ray photoelectron spectroscopy) system, equipped with a Shimadzu group Kratos Analytical Probe and an X-ray source (monochromatic (Al), 1486.6 eV). The work-functions of the ETLs were calculated via ultraviolet photoelectron spectroscopy (UPS) using an AXIS-ULTRA DLD spectrometer system, with source energy of 21.2 eV (He I). The top-view SEM images of ITO, SnO_2 , SnO_2 /IL and the cross-sectional image of a complete iOSC device was obtained from a field-emission scanning electron microscopy (FESEM) system (Hitachi-4700 and S4800). The energy dispersive X-ray spectroscopy (EDX) spectra and elemental mapping of SnO_2 and SnO_2 /IL ETLs were determined via EDX equipped with a FESEM system. The morphology of the ETLs as well as the photoactive layers was investigated using a Multimode-8 (Bruker, USA) with a tapping mode atomic force microscope (AFM), with a resonant frequency of 0.996 Hz. The photoluminescence studies for SnO_2 /P3HT:PC₆₀BM and SnO_2 /IL/

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