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Transition metal oxides on organic semiconductors

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ABSTRACT

Transition metal oxides (TMOs) on organic semiconductors (OSs) structure has been widely used in inverted organic optoelectronic devices, including inverted organic light-emitting diodes (OLEDs) and inverted organic solar cells (OSCs), which can improve the stability of such devices as a result of improved protection of air sensitive cathode. However, most of these reports are focused on the anode modification effect of TMO and the nature of TMO-on-OS is not fully understood. Here we show that the OS on TMO forms a two-layer structure, where the interface mixing is minimized, while for TMO-on-OS, due to the obvious diffusion of TMO into the OS, a doping-layer structure is formed. This is evidenced by a series of optical and electrical studies. By studying the TMO diffusion depth in different OS, we found that this process is governed by the thermal property of the OS. The TMO tends to diffuse deeper into the OS with a lower evaporation temperature. It is shown that the TMO can diffuse more than 20 nm into the OS, depending on the thermal property of the OS. We also show that the TMO-on-OS structure can replace the commonly used OS with TMO doping structure, which is a big step toward in simplifying the fabrication process of the organic optoelectronic devices.

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1. Introduction

Recently, transition metal oxides (TMOs) [1,2], such molybdenum trioxide (MoO_3) [3], tungsten oxide (WO_3) [4], vanadium pentoxide (V_2O_5) [5] and rhenium trioxide (ReO_3) [6], have gained great attention because of their wide applications in optoelectronic devices composed of organic semiconductors (OSs). For example, in organic light-emitting diodes (OLEDs) [7], they are used as anode modification interlayers [3], which can substantially reduce the hole injection barrier. They are also key components of charge generation layer in tandem OLEDs [8–10]. In organic solar cells (OSCs) [11], they are employed as charge extraction interlayer [12–14] and recombination layer [15,16]. These TMOs have many unique properties, such as high work function, semiconducting and good transparency, that are all very important for electrode interlayers and/or as charge generation/recombination materials. Among these properties, the high work function

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is the main reason for efficient operation of these TMO based functional layers. For example, thermal evaporated MoO_3 and WO_3 thin film have a high work function of 6.9 eV [17] and 6.7 eV [18], respectively, which are lying much lower than the highest occupied molecular orbital (HOMO) levels of most OSs and result in electron transfer from HOMO of OSs to conduction band (CB) of the TMOs (which is also termed as converting a hole current into an electron current) [1]. This process makes them popular choice to modify most electrodes (e.g., ITO [3], Au [19], Ag [20], Al [13], PEDOT:PSS [21,22] and graphene [23]) for efficient anodes in OLEDs and OSCs. This also makes them efficient p-type dopant. For example, with doping levels varying from intrinsic to high concentrations of up to 25 mol.% of MoO_3, the current density in 4,4′-N,N′-dicarbazole-biphenyl (CBP) based hole only device can be manipulated within a range of five orders of magnitude [24], which provides a direct evidence of p-doping by MoO_3.

Recently, inverted optoelectronic devices including inverted OLEDs [25], inverted quantum dot-LEDs [26] and inverted OSCs [13,27] have been extensively studied, owning to their ability to further improve the device stability compared with conventional structures. In these devices, TMOs have been widely employed due to their capability we mentioned above that they can modify almost all electrodes for efficient hole injection/extraction. The electrode modification effect in inverted devices is the same as that in normal devices. The difference between inverted and normal device structure is that, in the normal device structure, the hole transporting/extraction layer is deposited on TMO and in the inverted device structure, however, the TMO is deposited on the hole transporting/extraction layer instead. Interestingly, it is generally assumed that there is no difference between the OS-on-TMO and TMO-on-OS structures. However, we shall show it is not the case, at least in the system we studied.

Besides this, several studies [22,28,29] show that by introducing thin TMO film in the intermediate of OSs helps to improve the hole current and the cause was credited to the improved hole transport as a result of charge transfer (CT) complex formation at the interface between TMO and OS. For example, compared with ITO/NPB(40 nm)/Al structure, ITO/NPB(10 nm)/MoO_3(3 nm)/NPB(30 nm)/Al shows improved hole current [29]. The explanation of CT complex is based on the hypothesis that the OS and TMO thin films form a two-layer structure, where inter-diffusion between the OS and TMO is ignored. However, this inter-diffusion process between OS and TMO thin films sometimes can be underestimated, especially when depositing TMO on OS. Previous study indicates that, after depositing metal Li on OS, the diffusion depth of Li into OS can be up to 70 nm depending on the choice of the OS [30]. If similar process exists in TMO-on-OS structure, the mechanism for the improvement of the hole current should be revised. For example, in the case of ITO/NPB(10 nm)/MoO_3(3 nm)/NPB(30 nm)/Al, if the diffusion depth of MoO_3 into NPB is larger than 10 nm, the interface between ITO and NPB can be efficiently modified, which can also improve the hole current in the device.

In this paper, we have investigated the possibility of TMOs diffusion into OSs in the TMO-on-OS structure. It is shown that the diffusion process indeed exists. With MoO_3-on-NPB as a typical example, we show that the absorption of thin film of MoO_3-on-NPB is quite different from that of NPB-on-MoO_3. However, it is much similar to MoO_3 doped NPB thin film, indicating in the MoO_3-on-NPB structure MoO_3 is sufficiently mixed with NPB. By studying the current density–voltage (J–V) properties of a series devices with structure of ITO/NPB(x nm)/MoO_3(3 nm)/NPB($165 - x$ nm)/MoO_3/Al, we show that MoO_3 can diffuse into NPB up to 15 nm. We also studied MoO_3 with other OSs including 4,4′,4″-Tri(9-carbazoyl)triphenylamine (TCTA) and CBP, and the resulting diffusion depth of MoO_3 into TCTA and CBP is found to be up to 10 nm and 20 nm, respectively. By considering the thermal properties of NPB, TCTA and CBP, it is clear that the more thermal stable the OS is, the shorter depth the MoO_3 can diffuse into the OS. We also show that the diffusion of TMO into OS is good for simplifying the fabrication process of an organic electronic device. With OLEDs as an example, we employ MoO_3-on-CBP as hole transporting layer (HTL), the result device performance is found comparable to the device based on MoO_3 doped CBP HTL. This suggest that TMO-on-OS structure can replace the TMO doped OS structure, which eliminates the complex concentration control of the co-doping process in TMO doped OS structure. Hence, besides understanding the device physics, our finding could contribute to a more simplified organic device fabrication process.

2. Experimental

All devices were fabricated on commercial ITO-coated glass substrates. The ITO substrates were treated in order by ultrasonic bath sonication of detergent, de-ionized water, isopropanol and acetone, each with a 20 min interval. Then the ITO substrates were dried with nitrogen gas and baked in an oven at 80 °C for 30 min. Subsequently, the substrates were transferred into a thermal evaporator, where the organic, inorganic and metal functional layers were deposited sequentially at a base pressure lower than 4×10^{-4} Pa. The evaporation rates were monitored with several quartz crystal microbalances located above the crucibles and thermal boats. For organic semiconductors and metal oxides, the typical evaporation rates were about 0.1 nm/s and for aluminum, the evaporation rate was 1–5 nm/s. For the co-doping process, the rates of each material can be precisely monitored by the quartz crystal microbalances. The intersection of Al and ITO forms a 3 mm × 3 mm active device area. For the absorption measurement, the samples were prepared on quartz substrate, which also underwent the same washing steps as the ITO substrates. The absorption spectrum was carried out on a LAMBDA 950 UV/Vis/NIR Spectrophotometer. J–V and L–V data were collected with a source meter (Yokogawa GS610) and a luminance meter (Konica Minolta LS-110) with a customized Labview program. The lifetime study was performed in nitrogen filled glovebox.

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