

Full Length Article

Optimizing the combination of MoO₃ interface layer and low pressure plasma treatment on indium tin oxide (ITO) anodeYunlong Jiang^{a,b,c}, Yingjie Cui^c, Xi Cui^c, Yu Zhang^{b,*}, Liang Zhou^{c,*}, Yi Feng^a, Tieqiang Zhang^a^a College of Physics, Jilin University, Changchun 130012, People's Republic of China^b State Key Laboratory on Integrated Optoelectronics, College of Electronic Science and Engineering, Jilin University, Changchun 130012, People's Republic of China^c State Key Laboratory of Rare Earth Resource Utilization, Changchun Institute of Applied Chemistry, Chinese Academy of Sciences, Changchun 130022, People's Republic of China

ARTICLE INFO

Article history:

Received 8 April 2017

Received in revised form 22 May 2017

Accepted 31 May 2017

Available online 3 June 2017

Keywords:

Electroluminescence

Indium tin oxide (ITO) anode

Plasma treatment

Interface modification

ABSTRACT

In this work, we performed the investigation on surface modification of indium tin oxide (ITO) anode by depositing MoO₃ interface layer and treating with low pressure plasma. Experimental results revealed both low pressure oxygen plasma treatment and MoO₃ interface layer are efficient in facilitating the injection of holes, while both modifications increase the ITO surface roughness. Interestingly, the electroluminescent (EL) device with 3 nm MoO₃ layer displayed the highest EL performances, which were even higher than those of the device with both modifications. By optimizing the selection of gas, treatment time, and applied electric field intensity of low pressure plasma treatment, the combination of 3 nm MoO₃ layer and 2 min low pressure nitrogen plasma treatment at 4000 V m⁻¹ was demonstrated to be most efficient in depressing the unexpected leakage current without sacrificing the injection rate of holes.

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1. Introduction

Organic light-emitting devices (OLEDs) have attracted great interest throughout the world owing to their potential application in solid-state lighting and in full-color flat panel displays [1–5]. Although OLEDs have been developed prosperously in recent years, further improvements are still needed for widely commercial applications. Since gratifying achievement was received by the first used of LiF as interface layer between the cathode/electron transport layer (ETL), more and more groups put their attention on the interface of electrode and organic function layer. Anode/hole transport layer (HTL) is one of the most important interfaces, which determines the injection property of holes and the final EL performances [6,7]. However, the anode and HTL interfaces were unfriendly for the injection of holes in most instances. For example, indium tin oxide (ITO), which was the most commonly used anode in OLEDs due to its high conductivity and transparency, had the work function of about 4.7 eV [8]. The highest occupied molecular orbital (HOMO) level (−5.5 eV) of the hole transport material di-[4-(N,N-

ditolyl-amino)-phenyl]cyclohexane (TAPC), which was often used as HTL due to its high hole mobility ($1 \times 10^{-2} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$), is about 0.8 eV lower than that of as-grown ITO [9]. So the high energy barrier makes it difficult for holes to inject directly from ITO anode into HTL. In addition, ITO is highly hydrophilic with a heavily hydroxylated inorganic surface, while the organic HTLs (e.g., TPD and NPB) are mostly hydrophobic. This huge difference is disadvantageous for the growth and stability of the organic films deposited on ITO surface. Consequently, insufficient hole injection caused the unbalanced carriers' distribution and high operation voltage, thus the low EL efficiency and poor stability. Therefore, the development of efficient anode/HTL modification in OLEDs is very important in enhancing the device performances and accelerating the application of OLEDs.

Surface modification on ITO has been demonstrated to be an effective approach in improving the injection of holes and enhancing the device performances. In the past decades, many physical and chemical methods had been attempted to improve the ITO/HTL interface characteristics by modulating the work function of the ITO surface, smoothening ITO surface, and eliminating the disparity of ITO/HTL interface. By treating ITO surface with low pressure plasma [10], UV ozone [11], chlorination [12], or by coating the ITO with a buffer layer such as copper phthalocyanine (CuPc) [13], poly

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3,4-ethylene dioxythiophene: polystyrene sulfonate (PEDOT:PSS) [14,15], CHFx [16], V_2O_5 [17], MoO_3 [18,19], Al_2O_3 [20], and WO_3 [21], et al., significant improvements of EL performances had been achieved. Amongst these previous reports, low pressure plasma treatment attracted a lot interesting due to its effect on the cleaning of the ITO surface and the enhancement of hole injection. Besides it, the equipment cost of low pressure plasma treatment is relatively low which is favorable for the application in commercial use. Thus, the low pressure plasma treatment was chosen as one of the surface modification methods in this work.

MoO_3 was widely used as interfacial material since the firstly reported by Shizuo Tokito [22]. Previously, Ma et al. have realized the significant enhancement of power efficiency by 57% and the decrease of driving voltage by 40% in the MoO_3 //Bphen based devices [23]. Recently, Ma et al. adopted MoO_3 film as the interface modification layer on ITO surface to improve the interfacial stability and EL efficiency [24]. Although these methods could result in apparent enhancement of EL performances, EL mechanisms of these devices particularly the detailed hole injection processes are still unclear. Thus, further investigations are still needed.

In this work, we aimed to find a useful strategy for anode/HTL modification by utilizing low pressure plasma treatment as well as MoO_3 interface layer. By optimizing the combination of MoO_3 interface layer and low pressure oxygen plasma treatment on ITO surface modification, we found that 3 nm was the optimal thickness for MoO_3 interface layer. Interestingly, the device deposited with 3 nm MoO_3 film on ITO anode displayed even higher EL performances than that of the device modified with both 3 nm MoO_3 film and low pressure oxygen plasma treatment on ITO surface. Furthermore, the applied electric field intensity and treatment time of oxygen or nitrogen plasma were optimized in detail to better understand the effect of these modifications. Finally, 2 min low pressure nitrogen plasma treatment with 4000 V m^{-1} applied electric field was demonstrated to be most efficient in depressing the unexpected leakage current.

2. Experimental section

All the organic materials used in this study were obtained commercially and used as received without further purification. ITO coated glass with a sheet resistance of $10\ \Omega\text{ sq}^{-1}$ was used as the anode substrate. Prior to film deposition, patterned ITO substrates were cleaned with detergent, rinsed in de-ionized water and dried in an oven. And, some of the cleaned and dried ITO substrates were finally treated with oxygen or nitrogen plasma at a pressure of 10 Pa by adjusting the applied voltage and treatment time. All organic layers were deposited with the rate of 0.1 nm s^{-1} under high vacuum ($\leq 3.0 \times 10^{-5}\text{ Pa}$). The doping light-emitting layers (EMLs) were prepared by co-evaporating DBzA and host material from two individual sources, and the doping concentration was modulated by controlling the evaporation rate of DBzA. MoO_3 , LiF and Al were deposited in another vacuum chamber ($\leq 8.0 \times 10^{-5}\text{ Pa}$) with the rates of 0.01, 0.01, and 1.0 nm s^{-1} , respectively, without being exposed to the atmosphere. The thicknesses of these deposited layers and the evaporation rate of individual materials were monitored in vacuum with quartz crystal monitors. A shadow mask was used to define the cathode and to make ten emitting dots with the active area of 9 mm^2 on each substrate. Current density-brightness-voltage (J - B - V) characteristics were measured by using a programmable Keithley source measurement unit (Keithley 2400 and Keithley 2000) with a silicon photodiode. The EL spectra were measured with a calibrated Hitachi F-7000 fluorescence spectrophotometer. The external quantum efficiency of EL device was calculated based on the photo energy measured by the photodiode, the EL spectrum, and the current pass through the device. The

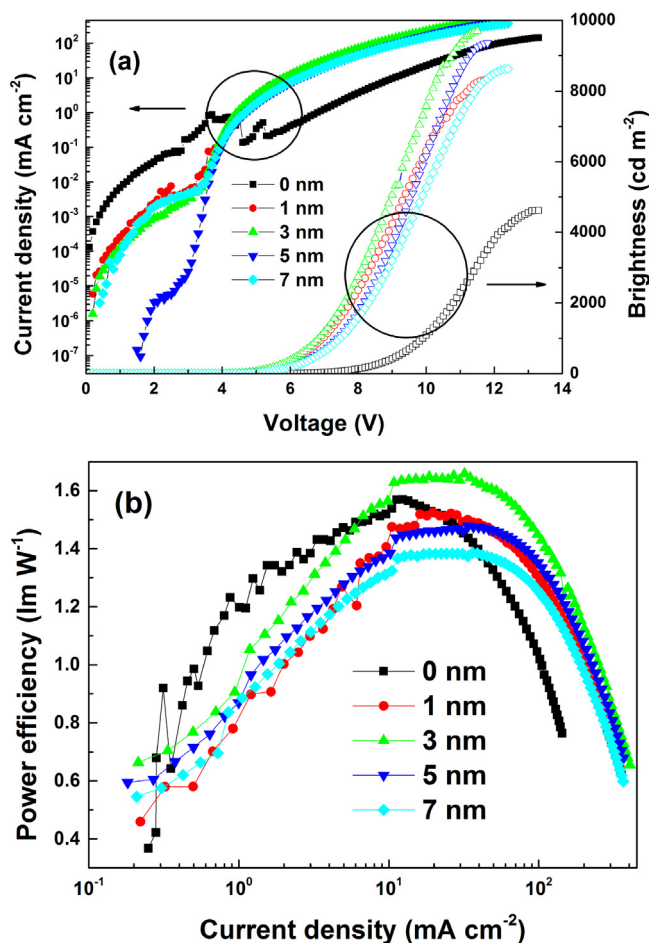


Fig. 1. (a) Current density-brightness-voltage characteristics of the devices based on ITO anode with different thicknesses of MoO_3 interface layer without low pressure oxygen plasma treatment. (b) Power efficiency-current density characteristics of the devices based on ITO anode with different thicknesses of MoO_3 interface layer without low pressure oxygen plasma treatment.

atomic force microscopy (AFM) pictures of the anode surface were obtained using a BRUKER Dimension Icon atomic force microscope.

3. Results and discussion

To optimize the thickness of MoO_3 interface layer, a series of EL devices with the structure of ITO/ MoO_3 (x nm)/TAPC (50 nm)/mCP (20 nm)/TmPyPB (40 nm)/LiF (1 nm)/Al (100 nm) were firstly fabricated and examined based on the ITO substrates without low pressure plasma treatment. Fig. 1(a) depicted the current density-brightness-voltage characteristics of these devices, while Fig. 1(b) depicted the power efficiency-current density characteristics of these devices. The key properties of these devices were listed in Table 1. Compared with the device based on ITO anode without any modification (defined as device A and reference device (RD)), these devices based on MoO_3 modified ITO anode displayed higher current density, higher brightness, lower operation voltage, but lower EL efficiencies. With increasing thickness of MoO_3 layer, current density and maximum brightness increase gradually to the highest values at 3 nm and then decrease slightly, while the turn-on voltage ($V_{\text{turn-on}}$, which was defined as the voltage on which the brightness of 1 cd m^{-2} was obtained) and the operation voltage decrease firstly to the lowest values at 3 nm and then increase gradually. Based on these results, the optimal thickness of MoO_3 layer was determined to be 3 nm, and this device (defined as device B) achieved the maximal brightness of 9699 cd m^{-2} , current efficiency of 3.95 cd A^{-1}

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