Investigation of top-emitting OLEDs using molybdenum oxide as anode buffer layer*

LIN Hui(林慧), **YU Jun-sheng**(于军胜)**, **and ZHANG Wei**(张伟)
State Key Laboratory of Electronic Thin Films and Integrated Devices, School of Optoelectronic Information, University of Electronic Science and Technology of China, Chengdu 610054, China

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A high-effective bottom anode is essential for high-performance top-emitting organic light-emitting devices (OLEDs). In this paper, Ag-based top-emitting OLEDs are investigated. Ag has the highest reflectivity for visible light among all metals, yet its hole-injection properties are not ideal for anodes of top-emitting OLED. The performance of the devices is significantly improved using the molybdenum oxide as anode buffer layer at the surface of Ag. By introducing the molybdenum oxide, the hole injection from Ag anodes into top-emitting OLED is largely enhanced with rather high reflectivity retained.
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Conventional organic light-emitting diode (OLED) includes a transparent conductive anode, typical indium tin oxide (ITO), covered by an organic multilayer for selective carrier transport to the emissive film and a thick layer of opaque metal[1-3]. Using this kind of device architecture, the emissive light is coupled out through the bottom contact, making on-chip OLED integration with silicon-based driver electronics rather impossible, especially for high-resolution full-color displays, so an active matrix addressing scheme is desirable. Therefore, it is imperative to develop an OLED which emits light from the top surface of devices.

Top-emitting OLED and transparent OLED can be fabricated on opaque substrates, such as Si wafers and stainless substrates[4-8]. They also permit use of more complicated pixel circuits in active-matrix OLED (AM-OLED) displays for higher display quality yet without sacrificing aperture ratios of pixels. For such top-emitting OLED, the bottom anode is crucial for achieving high luminance efficiency[9-11]. Among various metals, Ag has the highest reflectivity in the visible light range. However, Ag is generally not considered as an ideal hole-injecting anode for OLED, due to its rather low work function (4.3 eV) is not sufficiently high for effective hole injection[12]. Numerous methods have been proposed to enhance hole injection, such as capping with a metal with high work function or a metal oxide, using UV-ozone treatment, and inserting an ultra-thin insulator[8,13-16].

In this paper, by inducing a thin molybdenum oxide at the surface of the Ag anode, the hole injection from Ag into OLED is substantially enhanced yet with rather high reflectivity retained. The performance of the devices is significantly improved using the molybdenum oxide. In particular, the top-emitting OLED with 5 nm-thick MoO_3 as anode buffer layer shows excellent performance. With Alq as emitting layer and NPB as hole transfer layer, a maximum luminance of 2365 cd/m^2 at voltage of 20 V is achieved.

Prior to the deposition process, glass substrates are cleaned by sequential ultrasonic rinses in detergent solution, acetone, distilled acetone and deionized water, and finally dried in high purity nitrogen. The structure of top-emitting OLED is Ag/MoO_3/NPB/Alq/Mg:Ag/Alq, which is named as device A. The deposition apparatus is OLED-V organic multifunctional film system (Shenyang Vacuum, China), outfitted with quartz crystal oscillator in situ to detect deposition speed and film thickness. The organic materials are deposited onto substrate successively at a rate of 0.1−0.2 nm/s under the pressure of 3×10^{-4} Pa. An alloy of Mg:Ag (10:1) is deposited by simultaneous vacuum deposition from two separated sources at a rate of about 1.1 nm/s under the pressure of 3×10^{-3} Pa. The emitting area of device using a shadow mask is made to be 4 mm × 5 mm.

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** E-mail: jsyu@uestc.edu.cn

Luminance (L), bias voltage (V) and current density (J) characteristics of OLED are recorded by combining the spectrometer with a Keithley 2400 programmable voltage-current source. All measurements are performed at room temperature.

Molecular structures of Alq and NPB are shown in Fig.1. The OLED is made using a hole transfer layer of NPB and an emitting layer of Alq. Fig.2 shows the schematic structures of top-emitting OLED, transparent OLED and conventional OLED in our experiment. Top-emitting OLEDs of Ag/MoO_3/NPB/Alq/Mg:Ag/Alq are fabricated with the thickness of MoO_3 layer changing from 5 nm to 40 nm. To compare with other kinds of OLEDs, a transparent OLED with structure of ITO/NPB/Alq/Mg:Ag/Alq named as device B and a conventional OLED with structure of ITO/NPB/Alq/LiF/Al named as device C are also fabricated. ITO anode films are prepared on glass substrate by DC magnetron sputtering technique with the assistance of H_2O vapor[17].

Fig.3 shows the current density-voltage-luminance (J-V-L) characteristics of top-emitting OLED with different MoO_3 thicknesses. From Fig.3(a), it could be seen that with the increasing of MoO_3 thickness from 0 nm to 5 nm, the current density is changed significantly. The J-V curve of the top-emitting OLED without MoO_3 is almost linear, which indicates that this device does not have typical diode property. The reason may be that the pure metal Ag anode can not be injected efficiently to the device. The device with structure of Ag/MoO_3/NPB/Alq/Mg:Ag/Alq exhibits much higher current density than the device without MoO_3, and displays near Ohmic transport at low current density followed by the trap-free space charge limited current.

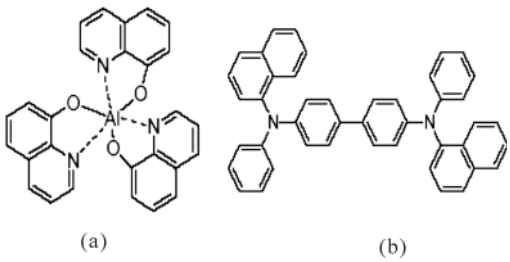

Fig.1 Molecular structures of (a) Alq and (b) NPB

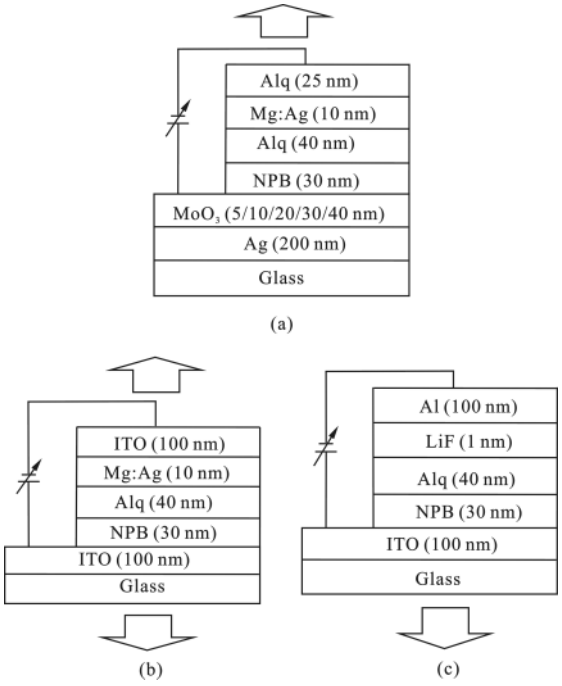

Fig.2 Device structures of (a) top-emitting OLED, (b) transparent OLED and (c) conventional OLED

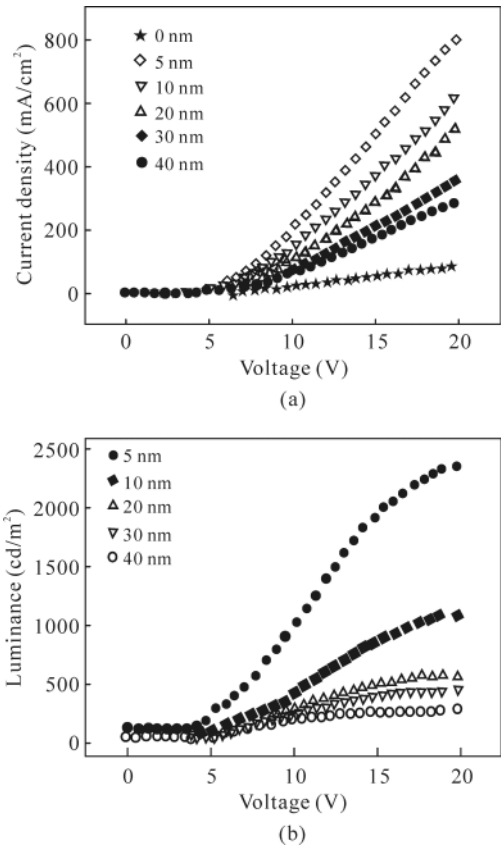

Fig.3(a) Current density–voltage characteristics and (b) luminance–voltage characteristics of top-emitting OLED with different MoO_3 thicknesses

Fig.3(b) shows the L-V characteristics of top-emitting OLED with different MoO_3 thicknesses. When the bias voltage increases, electron and hole injections are also enhanced, and devices begin to emit green light from Alq layer. It could be seen from Fig.3(b) that the maximum luminance for 5 nm-thick MoO_3 device is 2365 cd/cm^2 at driving voltage of 20 V. Compared with the current density in Ag/NPB/Alq/Mg:Ag/Alq device, the electron could be the only charge carrier, and no exciton is formed in the device, so no light is emitted out of the device. For the Ag/MoO_3/NPB/Alq/Mg:Ag/Alq

device, excitons ascribed to light emission are formed. The increase of luminance attributed to the proper MoO_3 thickness is suggested to facilitate the injection of the carrier into the emission zone and the electron-hole recombination. Accordingly, the MoO_3 film with the thickness of 5 nm is the optimum choice for the top-emitting OLED using MoO_3 as anode buffer layer.

The behavior of the buffer layer which is inserted on surface of Ag anode can possibly be understood as follows. Firstly, it is well known that MoO_3 is a wide-gap material with a band gap of 3.1 eV[18] and a superb hole injection material for anode[19], and the hole injection barrier from the metal anode to the HTL is lowered, so with the existence of the MoO_3, the performance of the top-emitting OLED is improved. It also can be further understood from the energy level diagram of device, which is shown in Fig.4. For the device with the MoO_3 buffer layer, the injection of holes from Ag into the hole transport layer becomes more effective due to the reduction of the Ag/NPB offset energy[20]. The significant enhancement of the hole injection current should be attributed to the low operational voltage and high luminance in top-emitting OLED[16]. Secondly, the top-emitting OLED studied in this work is sandwiched between the reflective bottom anode and the transparent top cathode, resulting in strong microcavity effects with the proper thickness of MoO_3. As reported previously, the out-coupling efficiency can be greatly enhanced owing to the microcavity effect[8,21]. Thirdly, the surface roughness of the bare Ag substrate is a few nanometers in height[13]. The existence of the MoO_3 layer not only smooths the interface but also increases the adhesion between layers, and thus results in a more even current flow in the device.

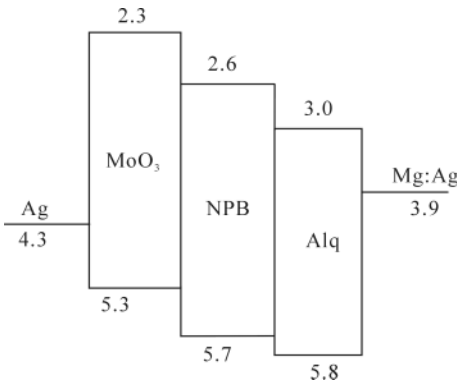

Fig.4 Schematic diagram of energy level for the top-emitting OLED with the MoO_3 buffer layer

Fig.5 plots the *L-V* characteristics of top-emitting OLED (device A), transparent OLED (device B) and conventional OLED (device C). From Fig.5, it could be seen that the luminances of devices A, B and C are 1096 cd/m^2, 1542 cd/m^2, and 4920 cd/m^2 at the same drving voltage of 12 V, respectively. It could be found that at the same voltage, the luminance of the device with metal anode is lower than that of the device with ITO anode. It indicates that the injection of holes from the ITO anode is more effective than that from the Ag/MoO_3 anode to the organic layer, which means there is still a lot of work to be done to improve the hole injection of metal anode.

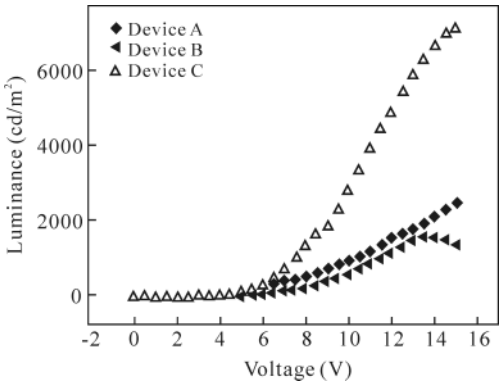

Fig.5 *L-V* characteristics of top-emitting OLED, transparent OLED and conventional OLED

In summary, we show that the hole-injecting properties of the Ag/MoO_3 anode can be effectively exploited to produce an efficient top-emitting OLED. By using an thermal evaporation MoO_3 buffer layer on the top of Ag anode, the light emission through the top of OLED is greatly enhanced, and it shows excellent luminance efficiency compared with the pure Ag anode structure. A top-emitting OLED with the luminance of 2365 cd/cm^2 at 20 V is processed.

From the analysis of the current density-voltage and luminance-current density characteristics of the top-emitting OLED devices, it is believed that the hole injection from the Ag anodes is greatly enhanced owing to the reduction of the injection barrier induced by the MoO_3. The energy band theory is applied to the Ag/MoO_3 interface to explain the experimental observations. These results represent a significant step for realizing high-resolution full-color displays based on top-emitting OLEDs.

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