Research on multi-color OLED with bi-directional charge transfer using Ag: Mg as intermediate electrode*

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(Received 16 December 2022; Revised 13 February 2023)

Organic light-emitting devices (OLEDs) have received widespread attention due to their excellent luminescence performance and flexibility. In this work, we demonstrate a novel two-color OLED, in which the forward and reverse units are vertically stacked together using Ag: Mg as an intermediate electrode for charge injection and transfer. Due to the extremely thin Ag: Mg film layer has a relatively smooth surface, it plays a critical role in the reverse unit light emission of the device. Thus, it is realized that the device emits green and red light when forward and reverse voltages are applied respectively. This provides a new possibility for future OLEDs lighting and display developments. The OLEDs were prepared by using such Ag: Mg intermediate electrodes, and a maximum brightness of 451.14 cd/m^2 and an external quantum efficiency (*EQE*) of 3.82% were obtained at a small current density.

Document code: A **Article ID:** 1673-1905(2023)06-0332-5

DOI https://doi.org/10.1007/s11801-023-2213-z

In recent years, organic light-emitting devices (OLEDs) have been developed rapidly[1-3]. OLEDs are widely used in lighting and display applications due to their high brightness, high color gamut, flexibility, and lack of backlighting[4-6]. White OLEDs are a popular candidate for next-generation lighting, replacing incandescent and fluorescent lamps[7,8]. In addition, light sources based on white OLED can be made flexible and transparent, providing new opportunities for architecture, visual art and diverse decoration.

Multi-color OLEDs have been attracting attention. Traditional OLEDs can only achieve monochromatic light emission, which significantly limits the development. The multi-color OLEDs can not only meet the demand for light color adjustable lighting but also improve the efficiency of the display and significantly reduce the cost. A lot of multi-color OLEDs have been studied in the laboratory[9-11]. Voltage-controlled color-tunable OLEDs are made using a mixture of small molecules and polymers[12,13]. By varying the voltage to excite organic materials with different spectral energy distributions, color-tunable OLEDs with multiple light colors can be achieved. The incomplete burst of low-bandgap materials prevents tuning to pure blue. This method has been shown to attain multi-color emission with pure color chromaticity[14]. The use of parallel or comb-like arrangement of two different light-emitting units in the same plane is also one of the methods to achieve multi-color tunable OLEDs[15,16]. Color-tunable OLEDs with parallel or comb-like arrangement in the same plane have contributed to the development of multifunctional displays[17], but they require additional structures in the manufacturing process and increase the cost. Multi-color tunable can also be achieved by vertically stacking two light-emitting units. This method requires the addition of intermediate electrodes in the two light-emitting units, usually using indium tin oxide (ITO)[18,19], Ag/Au[20], Ag[21], silver nanowire (AgNW)[22], etc, and requires a fine mask alignment process to achieve[23]. However, the fine mask alignment process is complex and expensive. Stacked OLEDs have a smaller pixel size and thus a higher fill factor compared to conventional parallel structures[24].

In this study, we demonstrate a stacked two-color OLED without fine mask alignment process to achieve multi-color luminescence. Without fine mask alignment process, the manufacturing cost will be greatly reduced, and the pixel size is expected to be smaller. The translucent metal electrode Ag: Mg is used as an intermediate electrode for charge injection and transfer, and the process complexity is reduced by eliminating the need to lead the intermediate electrode out. The extremely thin Ag: Mg has a good transmittance, and this translucency allows the deposition of two electrodes by stacking without affecting the device's luminescence. Since Ag: Mg has a good charge injection capability to provide bi-directional charge transfer, the device can achieve the emission of two light-emitting units, and the two light-emitting units do not affect each other. The positive unit emits green light when a forward voltage is applied

* This work has been supported by the Education Department of Shaanxi Province Serves the Local Special Plan Project (No.17JF006).
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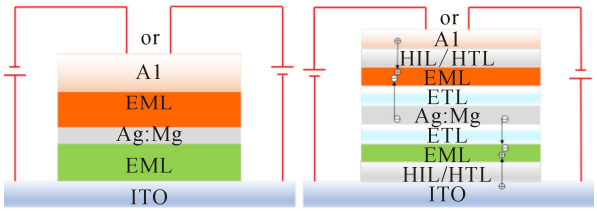
to the device, whereas the inverted unit emits red light when a reverse voltage is applied to the device.

We propose a new device structure, as shown in Fig.1, in which a very thin Ag: Mg is used as an intermediate electrode to connect the upper and lower different light-emitting units, without the need to adopt a delicate mask alignment process to lead out the intermediate electrode as the connection contact point. Ag: Mg is an excellent translucent electrode used in transparent devices in the past[25]. When the forward and reverse biases are applied separately, Ag: Mg can inject and transfer charge to the two light-emitting units respectively, thus enabling the two units to emit light independently.



EML: organic emissive layer; HIL: hole injection layer; HTL: hole transport layer; ETL: electron transport layer

Fig.1 Structure of multi-color OLED

First, ITO glass substrates were cleaned with continuous ultrasound using deionized water and ethanol, each for 10 min. After that, the cleaned ITO glass substrates were dried in an oven and treated in a UV-ozone instrument for 30 min. The ITO glass substrates were transferred to the thermal evaporation chamber. Organic layers and aluminum (Al) were sequentially deposited on the ITO glass substrates when the vacuum was lower than 4×10^{-4} Pa. Organic layers evaporation rate were 2 Å/s, the rate of evaporation for aluminum was 4 Å/s, and the rate of evaporation for molybdenum trioxide (MoO_3) was 0.2 Å/s.

The MoO_3 was used as the hole injection layer (HIL) due to the excellent hole injection ability of MoO_3. 1,1-bis[4-[N,N-di(p-tolyl)amino]phenyl]cyclohexane (TAPC) was used as the hole transport layer (HTL), because TAPC has an extremely high hole mobility while having electron blocking effect. 1,3,5-tri[(3-pyridyl)-phen-3-yl]benzene (TmPyPb) and ET20 were used as the electron transport layer (ETL), both of which have good charge transport ability. Fig.2 displays the structure of the devices and the energy band alignments. All organic materials were provided by Jiangsu Industrial Technology Research Institute (JITRI) without further purification.

The surface roughness was measured in tap mode using an atomic force microscope (AFM) (Dimension Icon, Bruker Corporation). Current density-voltage (*J-V*) and luminance-voltage (*L-V*) characteristics were measured using a Keithley 2400 source measurement unit. Transmittance and reflectance were measured using a UV spectrometer (PE Lambda 750s UV-Vis-NIR).

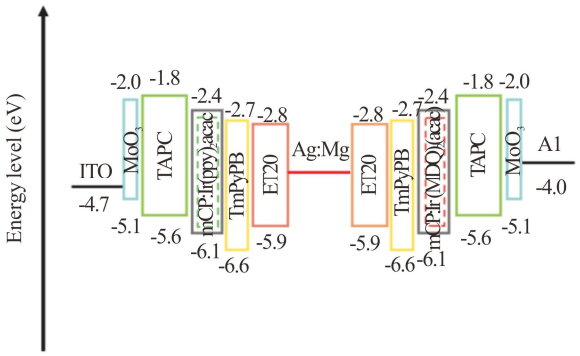


Fig.2 Energy level diagram

To intuitively acquire the result of Ag: Mg films, the AFM images are shown in Fig.3, where the ratio of Ag doped with Mg is different. Fig.3(a), (b) and (c) show the images of Ag: 5% Mg film, Ag: 8% Mg film, and Ag: 10% Mg film at 10 nm, respectively, with the root-mean-square surface roughness (R_{rms}) of 1.09 nm, 1.14 nm, and 1.19 nm. It can be seen that the roughness of the film changes with the increase of doping rate. It is obvious that surface roughness is a characteristic that affects the compatibility of transparent electrodes with devices. Compared with ITO image in Fig.3(d), Ag: Mg film has smoother surface and lower roughness than ITO, which indicates that the Ag: Mg film has better film forming property. In order to further evaluate the surface morphology of Ag: Mg films, scanning electron microscope (SEM) images of Ag and Ag: Mg films are shown in Fig.4. Fig.4(a) and (b) are SEM images of Ag and Ag: Mg respectively, which also indicate that the surface morphology of Ag: Mg film is smoother than that of Ag film. Small surface fluctuations will reduce the possibility of device breakdown and short circuit, and a uniform film layer is closely related to good performance.

Fig.5 shows the optical transmittance and reflectance characteristics of the Ag: Mg electrode. As shown in Fig.5(a), the Mg-doped films have a higher transmittance than the pure silver films without Mg doping, which is why Ag: Mg is usually used as a translucent electrode. In the range from 400 nm to 650 nm, Ag: 5% Mg showed the highest transmittance, followed by Ag: 8% Mg and Ag: 10% Mg and almost the same transmittance. Fig.5(b) shows the reflectance of the films with different ratios of Mg-doped with Ag. The reflectance of pure silver film and Ag: Mg film is opposite, while Ag: 5% Mg film and Ag: 8% Mg film have the same trend. Moreover, the reflectance of Ag: 8% Mg is low at light wavelength greater than 500 nm, which will affect the luminescence of the inverted unit in the stack structure.

Next, the structure of the prepared device is ITO/MoO_3 (5 nm)/TAPC (40 nm)/mCP: $Ir(ppy)_2acac$ (20 nm, 5%)/TmPyPb (10 nm)/ET20 (30 nm)/Ag: Mg (10 nm, 8%)/ET20 (30 nm)/TmPyPb (10 nm)/mCP: $Ir(MDQ)_2acac$ (20 nm, 3%)/TAPC (40 nm)/MoO_3 (5 nm)/Al (120 nm). Fig.6(a) shows the *J-V* characteristic curve of the device.

Combined with Tab.1, it can be seen that the current density of the device is different when the forward and reverse voltages are applied to the device, respectively, which may be related to the electron injection barrier. The turn-on voltage of device under forward and reverse biases is about 7.89 V and 8.57 V, respectively. Fig.6(b) shows the *J-L* characteristic curve of the device, it can be seen that even at low current densities, the maximum brightness of the forward and reverse units can reach about 451.14 cd/m^2 and 261.55 cd/m^2. Fig.7(a) shows the efficiency curves of the device. Combined with Tab.1, the maximum current efficiency (*CE*) and power efficiency (*PE*) of the device can reach 9.02 cd/A and 3.17 lm/W under forward bias, and the maximum external quantum efficiency (*EQE*) is 3.82%, which is by far the more impressive performance. Due to the difficulty of electron injection, the maximum current efficiency and power efficiency are 5.26 cd/A and 1.65 lm/W under reverse bias, and the maximum *EQE* of 3.12%. These results indicate that the OLED can be achieved by using a tandem structure with Ag: Mg interconnecting layers.

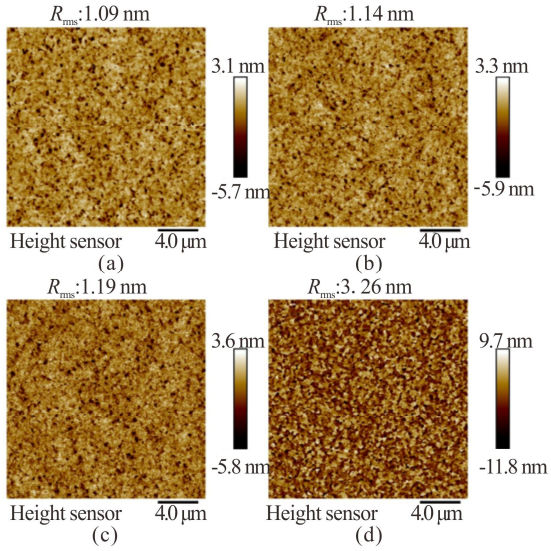


Fig.3 AFM topography images of (a) Ag: 5% Mg, (b) Ag: 8% Mg, (c) Ag: 10% Mg and (d) ITO

Fig.7(b) shows the spectral characteristics of the device, and it is found that the wave peak of the device at forward voltage is located at 568 nm and emits green light. In comparison, the wave peak of the device at reverse voltage is located at 615 nm and emits red light. Additionally, the additional peak in the green spectrum may be related to the microcavity effect. This demonstrates that the device can achieve dual-color light emission by changing the voltage polarity, which provides more possibilities for future multi-color tunable OLEDs.

In summary, we fabricated a novel stacked OLED structure to achieve dual-color luminescence by using Ag: Mg as the intermediate electrode for bi-directional charge transfer. Without the need for a fine mask alignment process to induce the intermediate electrode, which

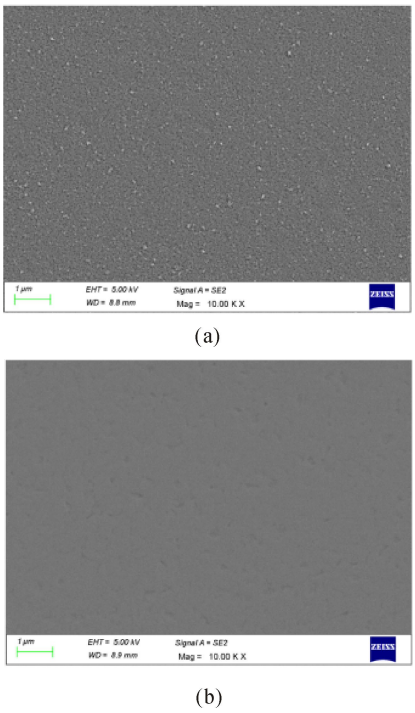


Fig.4 SEM images of (a) Ag (10 nm) and (b) Ag: 8% Mg (10 nm)

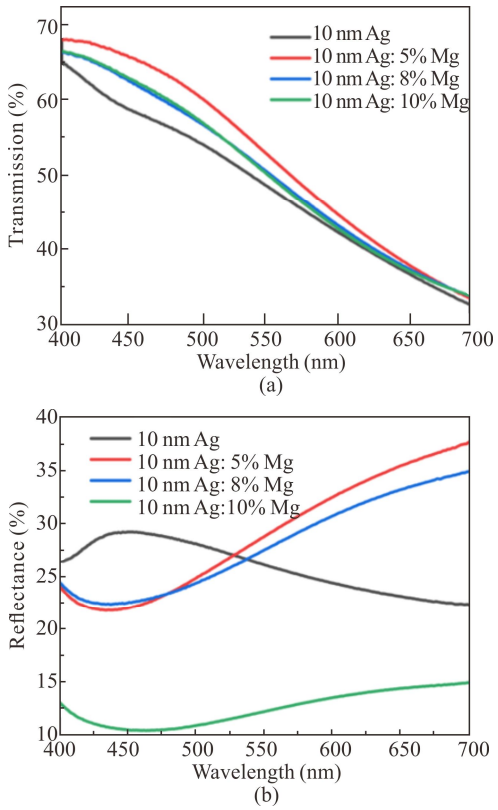


Fig.5 (a) Transmittance and (b) reflectance curves of films

not only simplifies the fabrication process and reduces costs, but also provides a new possibility to achieve smaller pixel size in the future. The 10 nm Ag: Mg films still have good surface morphology at different doping ratios, and the smooth film layer shows good device

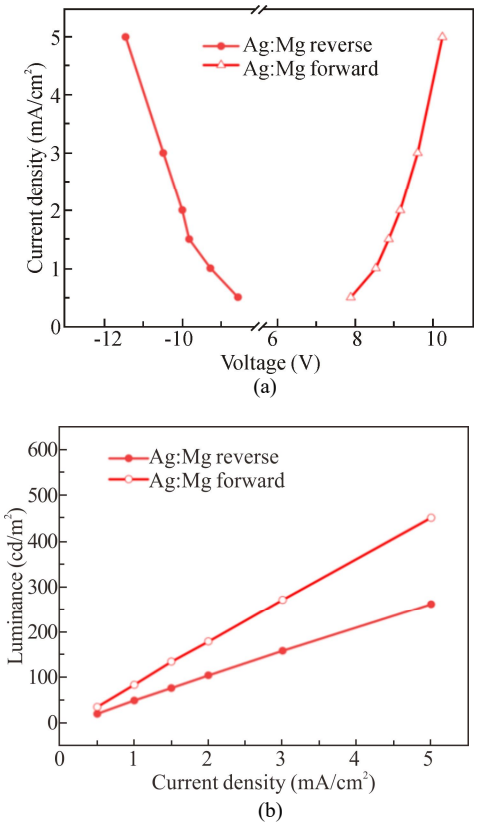
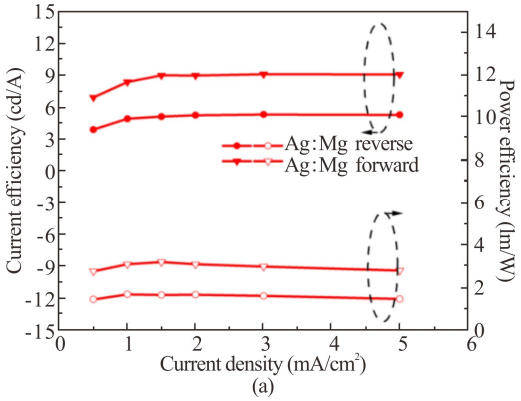


Fig.6 (a) *J-V* and (b) *J-L* characteristics of multi-color OLED

Tab.1 Summary of device performance for muti-color OLED

	V_{on} (V)	L (cd/m^2)	CE_{max} (cd/A)	PE_{max} (lm/W)	EQE (%)
Forward	7.89	451.14	9.02	3.17	3.82
Reverse	8.57	261.55	5.26	1.65	3.12



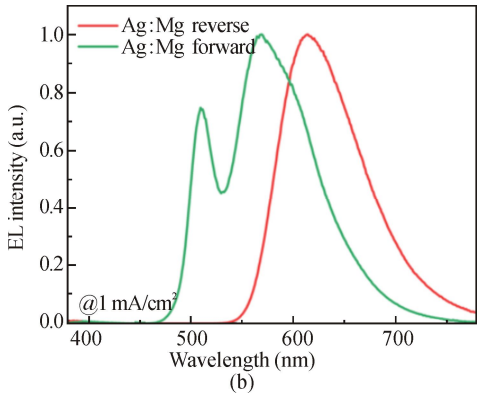


Fig.7 (a) *CE-J-PE* characteristics and (b) normalized EL spectra of multi-color OLED

performance. At the same time, the Ag: Mg films have good transmittance and can be used as an intermediate electrode to minimize the effect on the luminescence of the inverted unit. Finally, we prepared a device with Ag:Mg as an intermediate electrode connecting two stacked light-emitting units and tested the device's performance. A color-tunable OLED is fabricated with stacked green and red emitting units. The luminance of 451.14 cd/m^2 and the highest current efficiency and power efficiency of 9.02 cd/A and 3.17 lm/W are obtained at a low current density, and the maximum *EQE* is 3.82%, which is considerable for the efficiency of OLEDs with built-in electrode structure. The bi-directional charge transfer device does not require complex precision mask alignment nor is it voltage dependent. This device will be used to improve the future of multi-color lighting and display devices, while also providing more possibilities for the development of color temperature adjustable white OLEDs.

Ethics declarations

Conflicts of interest

The authors declare no conflict of interest.

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