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Efficient and Color-tunable Organic Light-emitting Diodes for Rear Light Application on the Motor Vehicle

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By designing double emissive layers, orange light-emitting layer (EML) and red EML and a space layer of blending hole-transporting material (HTM) with electron-transporting material (ETM), we have achieved efficient and color-tunable organic light-emitting diodes (OLEDs). It is found that the red shift of EL spectrum was enhanced when the ratio of HTM to ETM was 5:1. And a maximum current efficiency of 41.17 cd/A was reached. It has been demonstrated that two ultra-thin emissive layers could improve the current efficiency of the OLEDs. The current efficiency of the OLEDs with a 1 nm o;6range EML and a 1 nm red EML was enhanced by 27.4 % as compared to the reference devices. This is a novel approach to realizing voltage-controlled electroluminescence (EL) over different colors and intensity in a single OLED. The color-tunable OLEDs show the potential to be utilized as rear light on motor vehicle. We have investigated the effect of the ratio of HTM to ETM on the efficiency and spectral shift of the OLEDs through the control of recommendation zone.

Keywords: color-tunable organic light-emitting diodes, automotive lighting, space layer, recombination zone.

1. INTRODUCTION

Organic light-emitting diodes (OLEDs) have attracted general interest due to their characteristics such as flexible and self-luminous [1 – 6]. They have been applied successfully to the flat panel displays. Samsung and Huawei use OLEDs as the displays of their mobile phones. OLEDs have also made great progress in the fields of organic storage, organic optoelectronic, automotive lighting, etc. by optimizing the structure and organic materials of the devices [7 – 9]. Audi, BMW and many other car companies have applied OLEDs to their automotive lighting.

However, there are still some restrictions on the complete commercialization of OLEDs into automotive lighting, e.g. large-scale high-temperature stability and yield. In addition, the diversified development of automotive lighting has also attracted widespread attention. In the middle of 2006, the first car equipped with an intelligent Adaptive Front-lighting System (AFS) headlamp went into serial production and has, until now, generated a very positive reaction in the press all over the world. Shortly thereafter, an Adaptive Rear-lighting System with a signal light identical to the AFS principle used in various vehicles [10, 11].

In this study, we have developed an OLED device that could change from orange to red as the voltage changes. Orange light can be used as a stop light and red light as a turning signal lamp. The structure is simple in design. It achieves color-tunable and high performance just by adjusting the thickness of the space layer (SL) and the ratio of the hole-transporting material (HTM) to electron-transporting material (ETM) [12 – 18]. These materials are highly matched in energy levels, which can make the devices more efficient. The maximum current efficiency of the device is 41.17 cd/A. The maximum current efficiency of the OLED device with a 1 nm orange emitting layer (EML) and a 1 nm red EML can be increased by 27.4 % comparing to that with a 10 nm thick EML.

2. EXPERIMENTAL

The indium tin oxide (ITO) glass substrates having a sheet resistance of 10 Ω sq.$^{-1}$ were washed with deionized water, acetone and isopropanol, and then treated in an ultraviolet ozone atmosphere for 15 minutes before use. The devices were fabricated by conventional vacuum deposition of the organic layers and cathode onto an ITO coated glass substrate under a base pressure lower than 5×10^{-4} Pa. The typical deposition rates for organic materials were 0.6, 0.1 Å s^{-1}, and the typical deposition of aluminium (Al) was 5.0 Å s^{-1}.The two EMLs were Ga(pyimd)$_3$:8 wt.%PER53 and TCTA:8 wt.%EPY10. The host material Ga(pyimd)$_3$ was obtained from Lumtec Corp. (China). The other organic functional molecules purchased from e-Ray Optoelectronics Corp. (China). The structure of the devices and different ratios of the SL are shown in Fig. 1 a. The Fig. 1 b shows the chemical structures of TCTA and EPH31.

We used a Keithley 2400 source meter and a PR-650 Spectra Colorimeter to measure the current density – voltage-luminescence (J-V-L) characteristics. All measurements of the devices were carried out in an ambient atmosphere without further encapsulations.

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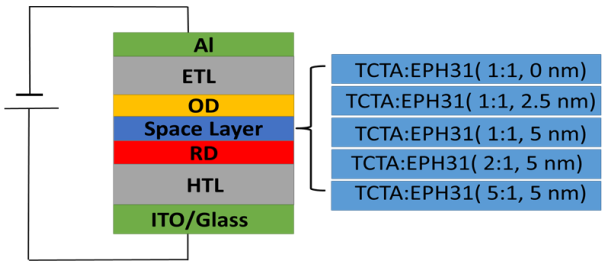

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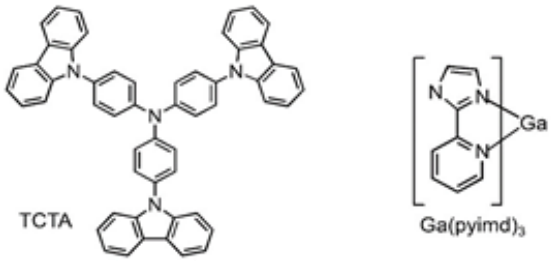

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Fig. 1. a – the structure of the OLED devices using space layers with different ratio of HTM and ETM; b – the chemical structures of the HTM and the ETM

3. RESULTS AND DISCUSSION

3.1. The effect of the ratio of HTM to ETM in the SL on the device performance

Fig. 2 a illustrates the current density (J) and luminance (L) characteristics against the applied voltage for the devices with different ratios of HTM to ETM and the 5 nm SL. The thin SL (≤ 5 nm) is used to balance the charge-transport of adjacent EMLs. By tuning the ratio of HTM to ETM and the thickness of the SL, an expected color shift from orange to red can be obtained.[14] It is clearly that the luminance of the device is the highest at the same voltage when the ratio of HTM to ETM is 1:1. Its turn-on voltage which is 2.6 V is the lowest. Due to the increased proportion of TCTA in SL, slightly higher turn-on voltage was observed. This is understandable from the triplet excited state of space interlayer material. Fig. 2 b is current density as a function of luminance, it can be found when the thickness of the SL is 5 nm, the device with HTM:ETM=1:1 had the highest current efficiency of 41.27 cd/A. It can be seen clearly in Fig. 2 that the greater the portion of TCTA with respect to EPH31, the lower current density and luminance devices exhibit. The facile SL can significantly enlarge the exciton recombination zone, and enhance the exciton harvesting from the interlayer and charge balance in the EML. It can be clearly observed that some amount of TCTA doped in EPH31 film significantly decreased the current density over the entire range of driven voltage. In particular, when the doping ratio of TCTA in EPH31 reached 5:1, the carrier transporting ability got worse. Accordingly, the hole/electron balance can be manipulated by adjusting the doping ratio of the TCTA: EPH31.

Fig. 2 c, d and e show the spectra and photographs of the devices with different ratios of HTM to ETM. Comparing these spectra and photographs, it can be found that the change of the peak position and color is obvious when HTM to ETM is 1:1. Fig. 3 summarizes the fitting spectra of devices with different ratios of HTM to ETM at different voltage. The EL spectra are black. The fitting spectra are red and superposed by two spectra in green.

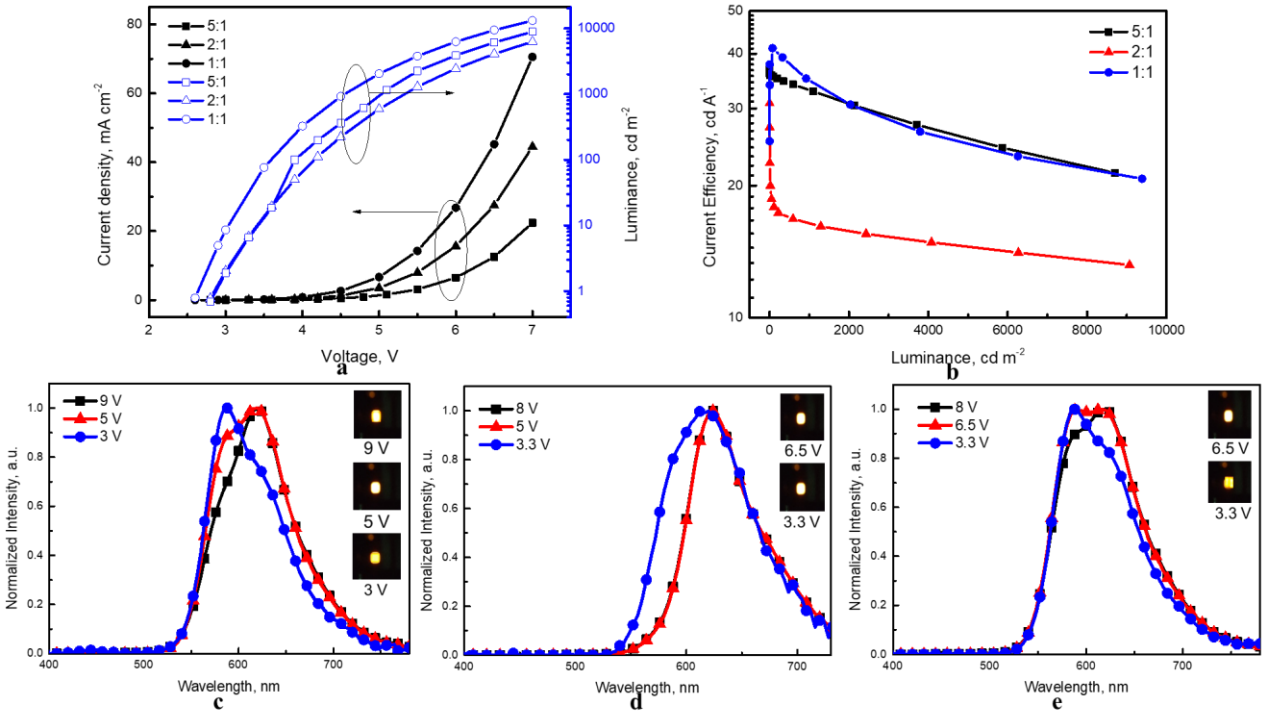

Fig. 2. a – the *J-V-L* characteristic; b – current density as a function of luminance of the devices with different ratios of HTM to ETM; c, d, e – the spectra of the devices at different voltages when the ratio of HTM to ETM is 1:1 (c); 2:1 (d); 5:1 (e). The thickness of the SLs of all devices is 5 nm. The insets are photographs of the devices

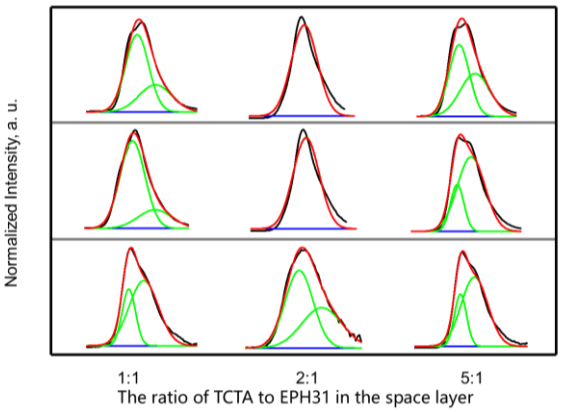

Fig. 3. EL spectra of the OLED devices with 5 nm SL before (black) and after (red) fitting. The ratio of HTM to ETM from left to right is 1:1, 2:1 and 5:1, respectively. The applied voltage from bottom to top is 3, 6 and 9 V, respectively

It is observed from the spectra of the device with HTM:ETM=1:1 that the peak position shifted from 586 nm to 612 nm, which indicates this device is of capability of tuning color. This means the exciton recombination zone shifted from an orange EML to a red EML side. The mixed SL plays a key role of channel in adjacent EML for electron/hole transfer, thereby leading to broadened exciton recombination zone covering the two EMLs.

3.2. The effect of the SL thickness on the device performance

Fig. 4 a illustrates current density (*J*) and luminance (*L*) characteristics against the applied voltage for the devices with 0, 2.5 and 5 nm SL, respectively. When the thickness of the SL was thicker than 5 nm, the turn-on voltage arose, and the power consumption increased too. [14] So we chose two devices with SLs thinner than 5 nm as reference devices. It can be seen from Fig. 4 a that when the ratio of HTM to ETM is 1:1, the brightness of the device with 5 nm SL is the highest. We can see from Fig. 4 b that the highest current efficiency is 41.17 cd/A when the SL thickness is 5 nm.

As the spectra and photographs show in Fig. 2 c, Fig. 4 c and Fig. 4 d, the device shows the most obvious color-tuning effect when the SL is 5 nm. This could be explained by the SL formation. When the thickness is only 2.5 nm, the SL may be not in the form of a thin film but in the form of islands. A thin film was formed until the SL thickness was increased to 5 nm.

3.3. The effect of the thickness of two ultra-thin emitting layers on the device performance

In order to explore the effect of the thickness of ultra-thin luminescent layers on device performance, we designed three devices with two EMLs of different thickness. Device 1-1 had 1 nm orange EML and 1 nm red EML, device 1-10 had 1 nm orange EML and 10 nm red EML, and device 10-1 had 10 nm orange EML and 1nm red EML [19–21]. Fig. 5 a and b illustrate the *J-V-L* characteristic and current density as a function of luminance. All the three devices have the same light which can be seen in the Fig. 5 a. The device with 1 nm orange and red EMLs has the highest luminance and current efficiency, which are 36110 lm/m^2 and 43.67 cd/A. However, the current efficiency of the device with the normal 10 nm EMLs is 34.27 cd/A.

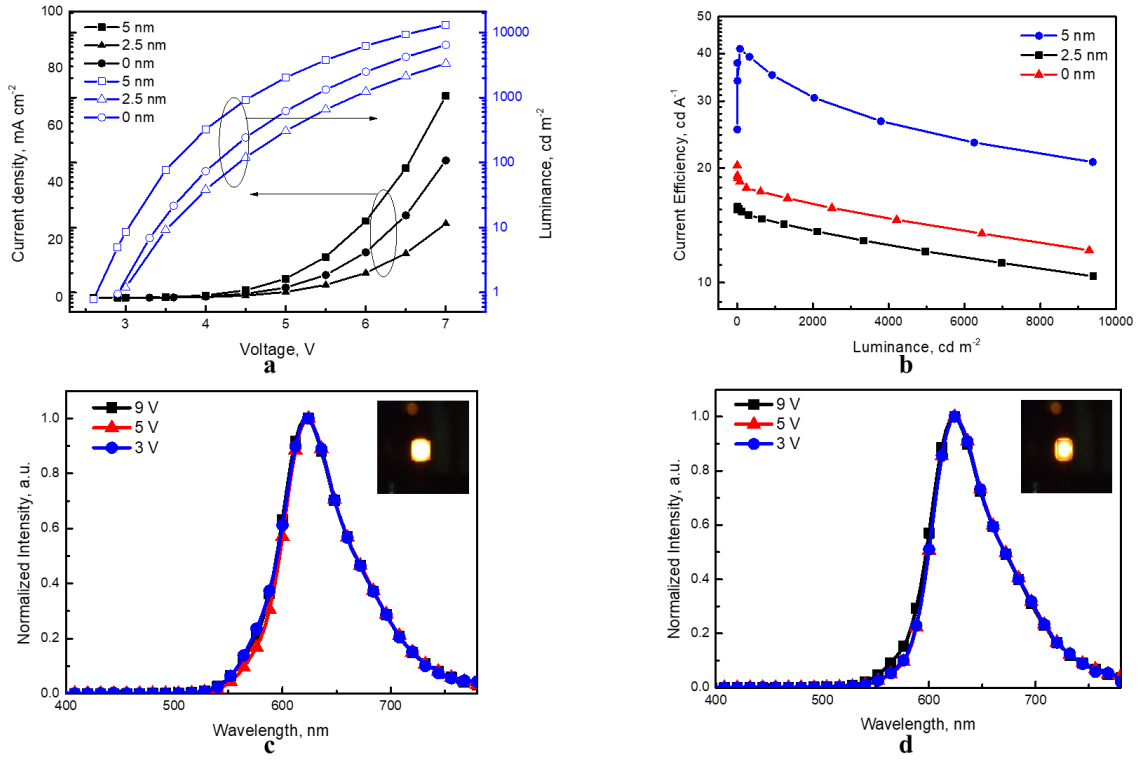

Fig. 4. a – J-V-L characteristics; b – current density as a function of luminance; c, d – the spectra and the photographs of the devices at 5 V. The SLs is 2.5 nm (c) and 0 nm (d). The ratio of HTM to ETM is 1:1

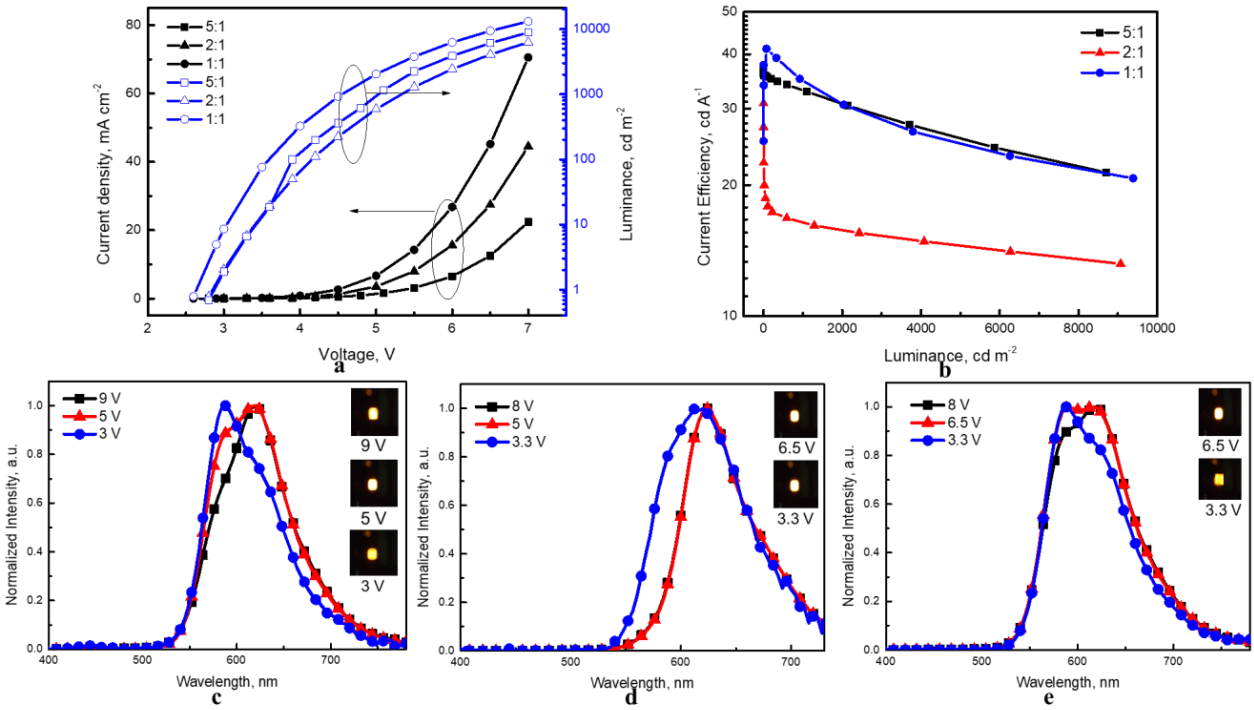

Fig. 5. a – the J-V-L characteristic; b – current density as a function of luminance; c, e – EL spectra of device 1-1 (c), device 1-10 (d) and device 10-1 (e)

Because the ultra-thin luminescent layer effectively reduces the annihilation of excitons, it results in higher efficiency.

We also explored the effect of ultra-thin emitting layer thickness on the color conversion performance of the devices. As shown in Fig. 5 c, d and e, the peak position of the EL spectra of the devices do not show clear shift. This suggests the EL spectra of the OLED devices are not sensitive to the thickness of the ultra-thin emissive layer, which could be attributed to the effective confinement of the excitons.

4. CONCLUSIONS

We have developed successfully high-performance and color-tunable OLED devices by optimizing the thickness of the SL and the ratio of HTM to ETM. A maximum current efficiency of 41.17 cd/A is achieved when the SL is 5 nm and the ratio of HTM to ETM is 5:1. In addition, it is found that the peak position of the EL spectra could shift from 586 nm to 612 nm by increasing the applied voltage from 3 V to 9 V when the ratio of HTM to ETM is 1:1. We demonstrate that the ultra-thin luminescent layer can improve the device performance significantly. These results provide a feasible idea of applying OLEDs in automotive tail-lamp.

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