Active emitting layer thickness dependence and interface engineering studies on the performance of DOPPP white organic light emitting diodes

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ABSTRACT A type of double light-emitting layer highly efficient white organic light emitting diodes (WOLEDs) with the stacked structures of ITO/NPB(30 nm)/Rubrene(0.2 nm)/DOPPP(x nm)/Bphen(40 nm)/LiF(0.6 nm)/Al(100 nm) have been fabricated by a vacuum thermal evaporation method. Wherein the fluorescent dye 5,6,11,12-Tetraphenylanthracene (Rubrene) and 1-(2,5-dimethoxy-4-(1-pyrenyl)-phenyl) pyrene (DOPPP) work as the yellow light layer and blue light layer, respectively. The performances of a series of developed devices are investigated by regulating the DOPPP thickness and tailoring its corresponding interlayer interfaces. The optimized device with DOPPP thickness of 20 nm demonstrates the best light emitting performance. The maximum brightness is 13910 cd/m² and the maximum current efficiency is up to 5.19 cd/A. With the increase of working voltage, the color coordinates migrate from (0.428, 0.433) to (0.382, 0.394), falling into the white light emitting scope. The light is changed from warm white light to neutral white light. When the thickness of DOPPP layer exceeds 20 nm, the efficiency and brightness tend to degrade slightly. The systematic investigations and complementary engineering studies upon the developed white WOLEDs also bring more opportunities to push the boundaries of current existing applications in an industrial level, e.g., flat panel display, human-machine interfaces lighting, and smart wearable devices etc

In recent years, organic light emitting devices (OLEDs) are mainly used for display and lighting source because of their large view angle, low-energy consumption and flexible display [1–19]. With the maturity of OLED technology and its wider application, OLED shows that the industry is growing rapidly and the market scale is growing very fast. Fig. 1 shows the global market size and forecast of organic light emitting diode (OLED). (Source: variant market research). Especially, the white organic light emitting diodes (WOLEDs) have attached the wide attention in lighting source since Kido [20] has successfully prepared WOLEDs for the first time in 1994. WOLEDs can exhibit the unique characteristics of flexible display when the substrate type is flexible. Therefore, many work have been done to obtain the highly efficient WOLEDs [21–34]. However, there are few organic dyes that emit the white light. Herein, the white OLEDs can be mainly fabricated by using colors. But, it is difficult to adjust the thickness of each luminescent layer and the order

between them in the preparation of white light devices which using three primary colors. It is easier to adjust when using the blue and orange light.

Doping technologies are widely used in the manufacture of nanomaterials and devices, expediting the development of all kinds of organic and inorganic materials and equipments [35–46]. Except for the fluorescent dye, many researchers have also used the methods of doping phosphorescent materials to prepare WOLEDs. However, the price of phosphorescent materials is relatively high and the doping concentration is difficult to be controlled. Although the performance of WOLEDs can be greatly improved, the preparation process is complex with high cost.

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1. Introduction

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Fig. 1. Global organic light emitting diode (OLED) market size and forecast, 2016–2024 (US \$ billion).

Moreover, the bandgaps of blue materials are so wider that the luminous efficiencies are lower relatively. Hence, it is very important to select the suitable blue fluorescent materials in the preparation of high performance WOLEDs.

In 2008, WU reported that DOPPP shows a high efficiency of the blue light fluorescent [42]. It is dipyrrenylbenzene derivatives. Its emission peak is shorter compared with the typical pyrene exciton. Melting point (T_m) and glass-transition temperature (T_g) are 336 and 135 °C, respectively. Its structure weakened the π - π stacking interaction between the molecules and improved the quantum efficiency of solid-state fluorescence. 5,6,11,12-Tetraphenylanthracene (Rubrene) is a mature and efficient orange fluorescent material. The yellow OLEDs and white OLEDs have achieved good performance by using Rubrene [47–53]. In our previous work, a group of non-doped white OLEDs with Rubrene as yellow layer and DOPPP as blue layer were designed and fabricated [54,55]. The performance of these devices was not very good, and the structure was relatively complex. On this basis, the structure of the device was further optimized. The hole injection layer m-MTDATA was removed and the hole transport layer NPB(N,N'-Bis(naphthalen-1-yl)-N,N'-bis(phenyl)-benzidine) was only retained. The hole barrier layer TAZ and the electron transport layer Alq3 were replaced by Bphen(4,7-Diphenyl-1,10-phenanthroline) and the luminescence layer was unchanged. The structures of the WOLEDs were optimized by regulating the thickness of blue layer (DOPPP). In this way, the device structure becomes simpler so that the device with better repeatability is easier to be prepared. Moreover, the prepared devices have better performance after optimizing the structure and they can be applied to warm white light devices for lighting.

2. Experimental

Fig. 2 demonstrates the chemical structures of two luminescent organic materials. In the devices, Bphen is used for electron transporting layer (ETL) and hole blocking layer (HBL). DOPPP acts as the blue light layer. NPB is used

for a hole-transporting layer (HTL). Rubrene acts as yellow light layer. All the purities of the organic materials are higher than 99.5%. The structure of the WOLEDs and energy level diagram of materials are showed in Fig. 3(a) and (b) respectively.

ITO glass is 20 Ω /sq. NPB and Bphen were bought from Jilin Aolaiide company. DOPPP, LiF and Rubrene were purchased from Xi'an Polymer Light Technology Corp. Firstly, the ITO-coated glasses were scrubbed clean. Then, they were immersed sequentially in ultrasonic baths of acetone, alcohol and deionized water for 15 min, respectively. Finally they were dehydrated in incubator at 140 °C. The four ITO glasses were placed in multivariate organic vapor deposition system. The system has eight heating sources and the ultimate vacuum is 1×10^{-5} Pa. The maximum temperature of the heating source is 800 °C. Organic materials and LiF are placed in five different crucibles, and the crucibles are placed on the heating sources respectively. According to the designed device structure, every organic material layer and LiF layer are evaporated in proper sequence. The evaporation rates of various materials were monitored by quartz crystal oscillators. The minimum rate it can monitor is 0.01 Å/s. Aluminum metal is placed in the tungsten wire basket, the evaporation rate is 15 Å/s. After the evaporation of Al, the temperature of heating sources are reduced. When the heat source temperature of LiF drops below 100 °C, the performance of devices can be measured. Electroluminescence (EL) spectra and CIE (Commission International de l'Eclairage) of the devices were gauged by PR655. The power was Keithley 2400. The measuring environment was at room temperature and air atmosphere.

3. Results and discussions

Fig. 4 illustrates the J-V characteristic curves of the devices. It illustrates that the current densities of devices A-D are almost the same when the driving voltage is low. With the increase of the thickness of DOPPP layer, the current density of the device begins to decrease. When the driving voltage exceeds 8 V, the current densities of device A are significantly higher than that of the others. It is attributed to the low conductivity of traditional organic materials used in the OLEDs. With the increase of the thickness of light emitting layer, the resistance of the device increases. According to Ohm's law, the current decreases with the same voltage. Ohmic conduction and trap limited conduction have the rectification characteristics of light emitting diodes [56,57]. It can be seen from Fig. 4 that they are the trap charge limited currents in the low voltage region [58]. When the voltage increases to a certain injection level, the traps are fully filled, so the ideal space charge limited currents are shown in the high voltage region.

Fig. 5 is the L-J characteristics of the devices. The graph shows that the luminance of the devices A-D are similar when the current densities are on the lower value. While the current densities are greater

Fig. 2. The chemical structures of the organic luminescent materials.

Fig. 3. The schematic structure of the white OLEDs (a) and respective energy level diagram of materials (b). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Fig. 4. The J-V characteristics of the devices.

shows a comparison of the E-J characteristics of the devices A-D. It that the current efficiencies of the devices increase rapidly when the densities are low. The change of efficiencies is relatively gentle when current density exceeds 1 mA/cm². The roll-off of efficiency is very The current efficiency of device B is higher compared to the others when current density is same. It is illustrated that the carriers in the light emitting layer are relatively balance when the thickness of DOPPP is 20 The efficiency starts to fall from device C (DOPPP is 25 nm). The efficiency is the lowest when DOPPP thickness is 30 nm (device D). The is that the recombination region of the excitons becomes wider with the increase of the DOPPP and the number of recombination of electrons holes increases. Therefore, the ratio of excitons participating in light emission increases and the efficiency of the device increases. The intermolecular π - π stacking effect worsens and reduces the current efficiency of the device when the thickness of the DOPPP is further increased. In addition, the working voltage of the devices rise and the efficiencies reduce with the increase of the blue light layer thickness increases. When the thicknesses of DOPPP continue to increase, the electron transport capacity of organic layer becomes weaker. The number of excitons in the DOPPP layer increases and the efficiency of blue light

Fig. 5. The L-J characteristics of the devices.

than 50 mA/cm², the brightness of the devices raises substantially. Moreover, the brightness of the four devices increases firstly and then decreases with the increase of the thickness of DOPPP under the same current densities. When the current density is high, the luminance of device B and C is higher more than that of device A and D. When the thickness of DOPPP is 20 nm (Device B), the luminance is the highest under the same current density. When the current density is 282.68 mA/cm², the maximum luminance of device B is 13910 cd/m². DOPPP is the material of subjective light emission [42]. Its hole-transport capability is strong. Therefore, the number of luminous exaction in DOPPP layer increases and the blue light enhance gradually when the thickness of DOPPP increases. The electrons which will be injected into the Rubrene layer are blocked by increasing the DOPPP thickness. The result is that the emission of DOPPP strengthens and the EL peaks become stronger. In addition, the characteristic of Rubrene is to capture holes and the main emission mechanism is direct exciton formation [59]. The electrons and holes will recombine to form excitons when the electrons are transported from the LUMO of DOPPP and injected into Rubrene layer. The excitons will generate yellow light through the radiative decay process.

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increases. The result is that the current efficiencies firstly increase and

Fig. 6. The E-J characteristics of the devices.

Fig. 7. The EL spectra of the devices A-D at 10 V. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

then decline under the same voltage. Therefore, the optimum thickness of the DOPPP layer is 20 nm and the maximum current efficiency is 5.19 cd/A.

Fig. 7 depicts the EL spectra of the devices A-D at 10 V. It depicts that the two main EL peaks of the four devices are found. They are the blue peaks of DOPPP and the yellow peaks of Rubrene, respectively. The main peaks of Rubrene are located at 560 nm and the shoulder peaks at 600 nm [49]. The thickness of Rubrene has not been changed, so its luminous intensity remains unchanged. The main EL peaks of DOPPP [42] are located at 452 nm (device A and B), 456 nm (device C) and 468 nm (device D), respectively. Moreover, their luminous intensities enhance when the DOPPP thickness increases. It is due to that the electrons can easily cross the DOPPP layer and enter the Rubrene layer when the thickness of the DOPPP layer is thinner (Device A and B). When the thickness of DOPPP increases, the number of electrons in the blue light layer increases. The result is that the number of the composite excitons increases and the blue light emission fortifies. At the same time, due to that the intermolecular π - π accumulation with the increase of the thickness of DOPPP, the blue main EL peak produces the redshift phenomenon that is from 452 nm to 468 nm. It can be seen

Fig. 8. The CIE coordinates of devices A-D at different voltages.

Table 1

The characteristics of devices A-D.

Device	Luminance _{max} (cd/m ²)	EL efficiency _{max} (cd/A)	CIE coordinates (Luminance _{max})	Color Rendering Index(CRI)
A	5 968	4.13	(0.391, 0.39)	63
B	13 910	5.19	(0.382, 0.388)	66
C	11 830	4.87	(0.379, 0.386)	67
D	6 855	3.34	(0.372, 0.384)	68

from Fig. 3b, the thickness of Rubrene is 0.2 nm. The HOMO and LUMO are 5.4 and 3.2 eV, respectively. The HOMO and LUMO of DOPPP are 5.9 and 2.8 eV, respectively. Therefore, a potential well structure is formed at Rubrene, where electrons and holes are easily captured and recombined to form excitons. The HOMO of Bphen is 6.4 eV, and the HOMO of DOPPP is 0.5 eV which is different from that of Bphen. Therefore, it is difficult for the holes to enter into the Bphen layer from the DOPPP layer, and most of them are limited at the interface between the DOPPP and Bphen. Bphen acts as a barrier to holes. The location of the blue light should be at the interface of DOPPP and Bphen.

Fig. 8 shows the CIE coordinates of devices A-D at different voltages. It can see that the light of the devices are all in the region of white. When the driving voltage is lower, the luminescence color of the devices A-D are all warm white light. With the raise of the voltage, the chroma changes from warm white to standard white light. The reason is that the blue light of DOPPP strengthens with the increase of the voltage and the blue and yellow light are relatively balanced. Therefore, the composite white light is near to the white center point. In addition the main luminance is Rubrene and the yellow light is stronger than the blue light. The blue luminous peaks produce the redshift with the increase of the blue layer thickness. In order to improve the chromaticity of the device, the method of strengthening blue ray emission is a good way to solve the above problem.

The characteristics of devices A-D are shown in Table 1. It can be seen that the four devices are all white OLEDs. The performance of device B not only the luminance but also the efficiency are all better than the others and the Color Rendering Index (CRI) is 66 at 1000 cd/m².

4. Conclusions

The double emission layer WOLEDs with the structures of ITO/NPB(30 nm)/Rubrene(0.2 nm)/DOPPP(x nm)/Bphen(40 nm)/ LiF(0.6 nm)/Al(100 nm) have been manufactured. The structure is non-doped. DOPPP and Rubrene are the blue and yellow emitting layer, respectively. The performance is optimized by regulating the thickness of DOPPP. When DOPPP thickness is under 20 nm, the light is mainly yellow. When the thickness is 20 nm, the performance is the best. The maximum brightness is 13910 cd/m² and the maximum current

efficiency is 5.19 cd/A. With the increase of driving voltage, CIE coordinates are changed from (0.428, 0.433) to (0.382, 0.388). The light is changed from warm white light to neutral white light and they are in the range of white light. The color rendering index(CRI) of four devices are A(63), B(66), C(67) and D(68) respectively when the luminance is 1000 cd/m². When the thickness of DOPPP layer is exceeded 20 nm, the performance the devices begin to fall.

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