

Article ID: 1007-2780(2010)05-0678-06

Effects of 4, 4', 4''-Tris[2-Naphthyl (Phenyl)Amino] Triphenylamine on Characteristics of Stacked White Organic Light-Emitting Devices Consisting of Separate Blue and Yellow Elements

YAN Hong-gang, ZHANG Fang-hui,
LU Jun-fu, JIANG Qian, LIU Ding-han

(School of Electric and Information Engineering, Shannxi University of Science and Technology,
Xi'an 710021, China, E-mail: bweia@qq.com)

Abstract: This paper systematically investigated the effect of the thickness of 2-TNATA on the characteristics of stacked white organic light-emitting devices consisting of separate blue and yellow elements. The device structure was 2-TNATA(x nm)/NPB(25 nm)/AND(30 nm):TBPE(2%):DCJTB (1%)/Alq₃ (20 nm)/LiF(1 nm)/Al(100 nm). According to the experimentation result, it was found the effects of thickness of 2-TNATA on carrier injection, color and thermal stability was very significant. The color of the white device can be adjusted by changing the thickness of 2-TNATA inserted. The device using 20 nm 2-TNATA as a hole injection layer showed pure-white light emitting with 1931 CIE chromaticity coordinates $x=0.319\ 7$, $y=0.349\ 6$, and the luminance of 12 230 cd/m².

Key words: 4, 4', 4''-tris[2-naphthyl (phenyl)amino]triphenylamine; white organic light-emitting devices; luminance characteristics

2-TNATA 对蓝与黄二基色分离的堆叠式白色有机发光器件性能的影响

阎洪刚, 张方辉, 陆君福, 蒋 谦, 刘丁菡

(陕西科技大学 电气与信息工程学院, 陕西 西安 710021, E-mail: bweia@qq.com)

摘 要: 研究了 2-TNATA 厚度对蓝与黄二基色分离的堆叠式白色有机发光器件性能的影响。器件结构为: 2-TNATA(x nm)/NPB(25 nm)/ADN(30 nm):TBPE(2%):DCJTB(1%)/Alq₃ (20 nm)/LiF(1 nm)/Al(100 nm)。根据实验结果, 2-TNATA 的厚度对载流子的注入、色稳定性、热稳定性影响明显。发光器件的颜色可以通过改变加入的 2-TNATA 层的厚度来改变。这种器件使用 2-TNATA 作为空穴注入层显示出了色纯度高的白光发射, CIE 色坐标 $x=0.319\ 7$, $y=0.349\ 6$, 亮度能够达到 12 230 cd/m²。

关 键 词: 2-TNATA; 白色有机发光器件; 发光特性

中图分类号: TN 383⁺.1 **文献标识码:** A

1 Introduction

White organic light-emitting diodes (WOLEDs) have attracted a great deal of attention because of their potential use in various applications, such as full color display, back-lighting, and also as candidates for the next generation lighting^[1]. In present, many methods are applied to improve the characteristics of WOLEDs. Multi-emitting layer WOLEDs have much higher efficiency than that of the single emitting layer devices, but the CIE(*x*, *y*) coordinates are often dependent upon drive current density, which is due to the shift of the exciton-recombination zone^[2-4]. It is a challenge to develop devices with stable color coordinates with respect to the bias voltage or current density^[5]. High color and stable efficiency have been got in three color hybrid white organic light emitting diodes^[6]. Hybrid type two color devices could also give stable color performances over a wide luminance range^[7].

In this work, stacked white organic light-emitting devices consisting of separate blue and yellow elements were fabricated. Especially, no blocking layer was inserted into the device in order to low the initial voltage, and 4, 4', 4''-tris[2-naphthyl (phenyl) amino] triphenylaminef (2-TNATA) was used as buffer layer of the device. We have mainly investigated effects of the thickness of 2-TNATA layer on characteristics of the white devices.

2 Experiments

The structure of these devices were constructed as follows: ITO/2-TNATA (*x* nm)/NPB (25 nm) / ADN (30 nm) : TBPE (2%) : DCJTB (1%) / Alq₃ (20 nm) / LiF(1 nm) / Al(100 nm).

The thickness of TBPE and DCJTB had been optimized. In the configuration, 2-TNATA was utilized as a hole injection layer(HIL), and N, N'-Bis-(naphthalen-1-yl)-N, N'-bis(phenyl)

ben-Zidine (NPB) was a hole transporting layer (HTL), 9,10-di-(2-naph-thyl)anthracene(ADN) was a blue emitter, tris (8-hydroxyquinoline) aluminum(III) (Alq₃) was the electron transporting layer(ETL) and green emitter, 4-(Dicyanomethylene)-2-t-butyl-6-(1, 1, 7, 7-tetramethylj-ulolidyl-9-enyl)-4H-pyran(DCJTB), 2, 5, 8,11-tetra -butyl - perylene (TBPe) were red and blue dyes, respectively.

The configuration and energy level diagram of the devices were shown on Fig. 1. The devices with different thickness of 2-TNATA were fabricated under the same deposition conditions included deposition rates, the pressure of the chamber, rolling rates of the glass plates and deposition temperature. Indium Tin Oxide (ITO) coated glass substrates were cleaned successively utilizing acetone and ethanol in an ultrasonic bath for 5 min, then were dried in the dryness ark. The surface of glass substrate was treated with air plasma before these organic layers were deposited on the glass substrates. Finally, organic layers 2-TNATA, NPB, EML, Alq₃ and compound cathode LiF/Al were thermally deposited on the substrates treated at a

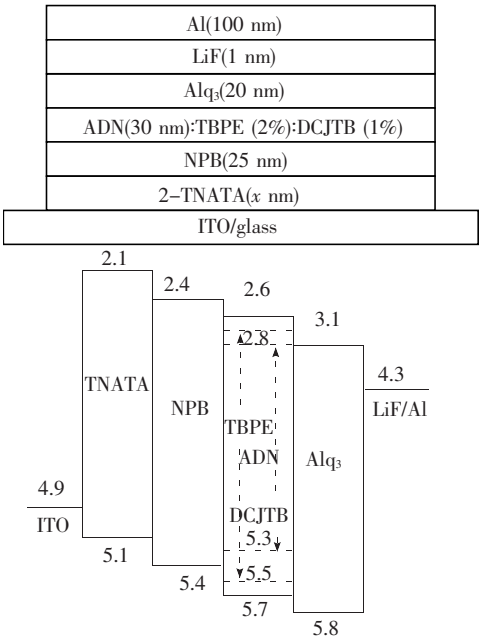


Fig. 1 Configuration and energy level diagram of the organic light-emitting devices

rate of 0.3 nm/s and 1 nm/s when the pressure of the chamber reached 6×10^{-4} Pa, especially, the composite film (EML) was prepared from simultaneous co-deposition of host and dopants from three separate sources. The active area of the devices was 210 mm², which was confined by a shadow mask with 14 mm × 15 mm openings. The evaporating rates and thickness of the organic and metal layers were monitored using quartz oscillators. All the parameters of the devices fabricated were measured at room temperature.

3 Results and Discussion

Fig. 2 shows the dependence of current density and luminance on different voltages. It can be seen from Fig. 2(a) and (b) that current density and luminance of the devices get higher as the drive voltages increasing. We illustrate the phenomenon like that charge carrier injection ability get stronger when the drive voltage is enhanced. At the same voltage, the phenomenon that current density of the device gets larger at

the beginning, then gets smaller as increasing of thickness of 2-TNATA can be observed.

We can find that the holes can be easily injected into the hole transporting layer if the buffer layer is thin (0~25 nm), in that condition, thin 2-TNATA buffer layer lows down carrier injection potential barrier which is higher about 0.5 eV between ITO and NPB. When the thickness of 2-TNATA is 30 nm, current density and luminance get rapidly smaller at the same voltage. Referring to Fig. 2 and Table 1, initial voltages of the devices with different thickness of 2-TNATA are nearly equal around 4 V except the device with 2-TNATA (30 nm) whose initial voltage is 4.68 V, however, initial voltages of the devices without 2-TNATA is 5.51 V. The injection barriers which is between ITO and NPB is 0.5 eV, it can be reduced to 0.3 eV after inserting 2-TNATA, so we get comparatively low initial voltage. At the same time, luminance, η_i (luminance efficient of the device), η_p (power efficiency) are obviously advanced after inserting the buffer layer of 2-TNATA. Thermal decomposition temperature of the device is enhanced to 60 °C because of existent 2-TNATA. T_g (glass transition temperature) of 2-TNATA commonly used as hole transporting materials is 110 °C. It is well-known that the higher the T_g of a material, the better is the morphological stability of an organic thin-film^[8]. From Table 1, performance of the device with 2-TNATA(20 nm) is the most excellent in all devices whose η_i can reach 4.24 cd/A at a current density of 1.43 mA/m², and highest luminance of the device is 12 230 cd/m² at a voltage of 13.0 V. η_i and highest luminance are low. In the structure of devices, the thickness of NPB, ADN and Alq₃ was optimized, what led to low luminance efficient. At a high voltage, some small spots would come forth in the active area of the devices. The devices would be penetrated at the place where the spots generated, so the luminance was low. Here we think the reason of spots come forth is that glass

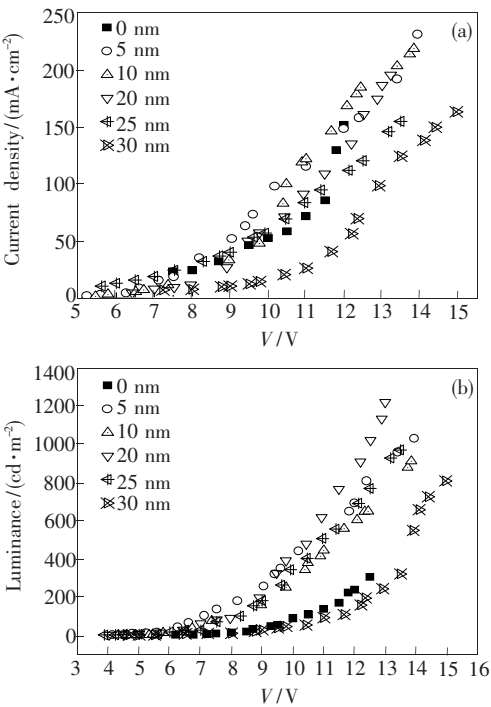


Fig. 2 Current density-voltage curves (a) and luminance-voltage curves (b) of the devices with different thickness of 2-TNATA

Table 1 EL performance of the devices with different thickness of 2-TNATA						
2-TNATA (<i>x</i> nm)	<i>V</i> _{turn on} (V)	Luminance ^{bc} (cd/m ²)	η _b ^d (cd/A)	η_p ^a (lm/W)	<i>T</i> ^b (°C)	CIE(<i>X</i> , <i>Y</i>) ^a
0	5.51	3 050 (12.5)	0.58(67.14)	1.261	31	(0.406 1,0.421 7)
5	4.12	10 340 (13.94)	4.77(4.28)	2.671	79	(0.342 8,0.380 9)
10	4.33	9 130 (13.84)	4.29(11.43)	2.291	59	(0.292 3,0.341 3)
20	4.03	12 230 (13.0)	4.24(1.43)	2.551	47	(0.319 7,0.349 6)
25	3.87	9 730 (13.49)	1.91(56.67)	2.810	43	(0.321 3,0.344 0)
30	4.68	8 100 (14.96)	1.13(137.61)	2.011	44	(0.289 8,0.312 9)

^a Values collected at 28.6 mA/m²; ^b Maximum values; Values in parentheses are the voltages ^c or current density ^d at which the maximum values were obtained.

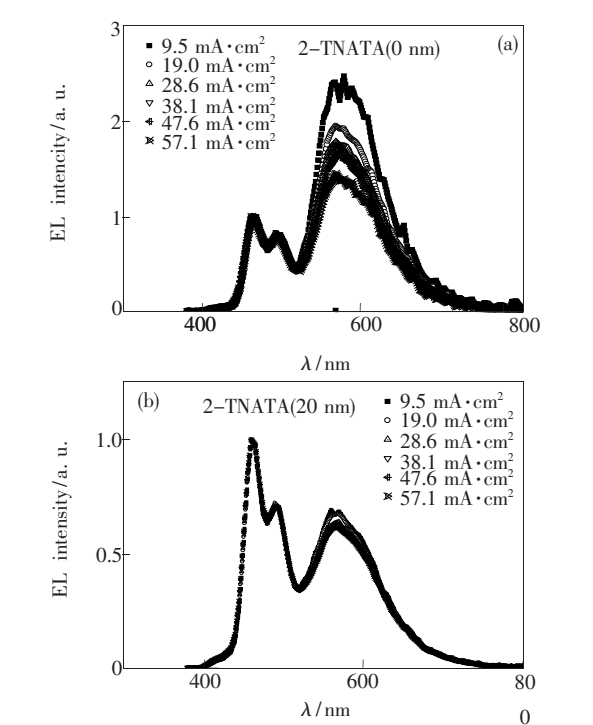


Fig. 3 EL spectra of the devices with 2-TNATA (0 nm and 20 nm) at different current density

substrates had not been cleaned perfectly, and clean methods was not best.

Fig. 3 shows the EL characteristics of the device with 0 nm and 20 nm 2-TNATA at different current density. In order to check which part of the emission increases quickly, the emission spectra at different current density are normalized. The fluorescent characteristics emission peaks of TBPE are at 459 nm and 480 nm, and characteristics emission peak of ADN is at

460 nm^[9-10]. Two EL blue peaks (460 nm and 490 nm) which can be seen from the EL spectra generate from interacting between ADN and TBPE. The yellow peak which come from Alq₃ (green) and DCJTb (red) is used as the other element of the white device. Three peaks value happen to slightly shift when the current density is enhanced after inserting 2-TNATA and it is shown that the changing of the current density slightly affected the emitting region of the devices. During the experiment, we test all CIE coordinates chromaticity of the devices with different thickness of 2-TNATA at different current density. We find that color changing lightly at different current density. It indicates that inserting of 2-TNATA have upgraded the color stability of the device. Simultaneously, it can be shown that the yellow band is alleviated as increasing of current density from Fig. 3, but this scale is not enough to make luminescence color obviously changing even if the device is without 2-TNATA. The CIE coordinates(*x*, *y*) of the devices without 2-TNATA are (0.430 2,0.438 9), (0.413 6,0.427 2), (0.406 1,0.421 7), (0.402 6,0.418 6), (0.398 2,0.416 5), (0.3909, 0.4807) at six different current density. Nevertheless, the color shift is not more than 0.02 units on the 1931 CIE chromaticity diagram. The color stability of the devices gets more excellent after inserting 2-TNATA layer.

Yellow peak of the devices falls off quickly, at the same time, color shift is visible at different thickness of 2-TNATA what are shown in Fig. 4(a) . The accurate CIE coordinates can be seen from Table 1. The luminescence color is nearly yellow at the beginning, then it turned to white significantly when 2-TNATA layer is inserted into the device. At the same current density, the luminance value rises rapidly. This indicates that the radiation exciton increased in the carrier combination zone. 2-TNATA is helpful to optimize the ability of carrier injection because it makes the number of holes injected into the device to get larger. After inserting 2-TNATA, holes become overmuch carrier which can be controlled by changing the thickness of 2-TNATA to affect combination zone of carriers. The device with 2-TNATA of 20 nm shows CIE coordinate (0.319 7,0.349 6), and the one which have 2-TNATA of 25 nm shows CIE coordinate (0.321 3, 0.344 0). They show a excellent color purity what is well adjacent to the CIE coordinate (0.33,0.33) . From Fig. 4 (a), we can see that yellow band is not only weakened, but also blue band (490 nm) of TBPE, simultaneously, what is not favorable to color shift from yellow to warm white.

From Fig. 4 (b) T - V characteristics, we can know that thermal stability of device is well below 9 V. We know that the luminance of the device can reach 2 000 cd/m² below 9 V, which is enough to satisfy the commercial need. In this diagram, we can also see hole injection ability is advanced after inserting 2-TNATA layer. Temperature of the device risen rapidly, what can be explains like that hole injecting is enhanced after inserting 2-TNATA layer. Increasing of injection holes leads to the tempera-ture risen rapidly, and the injection number of holes will be diminished as increasing of the thickness of 2-TNATA.

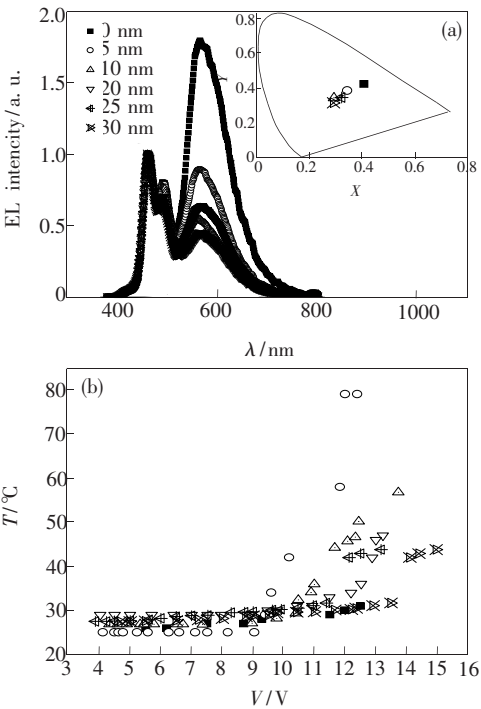


Fig. 4 (a)EL spectra of the devices with different thickness of 2-TNATA at 28.6 mA/cm², the inset shows the corresponding CIE coordinates; (b) Temperature *vs.* voltage curves of the device.

4 Conclusion

In conclusion, thickness changing of 2-TNATA inserted can affect on performances of the stacked white organic light-emitting devices consisting of separate blue and yellow elements the device. Adjusting thickness of the device, we can get the luminance 12 230 cd/m², and CIE coordinate(0.319 7, 0.349 6) can be obtained when thickness of 2-TNATA of the device is 20 nm. In addition, inserting of 2-TNATA can de-base initial voltage, enhance color stability and thermal stability of the white organic light-emitting devices at different current density. These can be ascribed to the number of holes injected being adjusted by 2-TNATA.

References:

[1] Guo F W, Ma D G. White organic light-emitting diodes based on tandem structures [J]. *Appl. Phys. Lett.* , 2005, 87(17):173510(1-3).

[2] Wu Y S, Hwang S W, Chen H H. Efficient white organic light emitting devices with dual emitting layers[J]. *Thin Solid Films*, 2005, 488 (1-2):265-269.

[3] Cheon K O, Shinara J. Combinatorial fabrication and study of dopedlayer-thickness-dependent color evolution in bright small molecular organic light-emitting devices [J]. *Appl. Phys. Lett.* , 2003, 83(10):2073(1-3).

[4] Cheon K O, Shinara J. Bright white small molecular organic light-emitting devices based on a red-emitting guest-host layer and blue-emitting 4,4'-bis(2,2'-diphenylvinyl)-1,1'-biphenyl [J]. *Appl. Phys. Lett.* , 2002, 81(9):1738 (1-3).

[5] Chen W B, Lu L L, Cheng J B. Characterization of two-emitter WOLED with no additional blocking layer [J]. *Optik*, 2010, 121(1):107-112.

[6] Yook K S, Jeon S O, Joo C W, *et al.* High efficiency, color stability, and stable efficiency roll off in three color hybrid white organic light emitting diodes [J]. *Appl. Phys. Lett.* , 2008, 93(7):073302(1-3).

[7] Wang Y, Hua Y L, Wu X M, *et al.* Performance enhancement of white-electrophosphorescent devices incorporating a mixed-transition layer [J]. *Appl. Phys. Lett.* , 92(12):123504(1-3).

[8] Gao Z Q, Xia P F, Lo P K, *et al.* p-doped p-phenylenediamine-substituted fluorenes for organic electroluminescent devices [J]. *Organic Electronics*, 2009, 10:666-673.

[9] Mi B X, Gao Z Q, Lee C S, *et al.* Reduction of molecular aggregation and its application to the high-performance blue perylene-doped organic electroluminescent device [J]. *Appl. Phys. Lett.* , 1999, 75:4055(1-3).

[10] Shi J M, Tang C W. Anthracene derivatives for stable blue-emitting organic electroluminescence devices [J]. *Appl. Phys. Lett.* , 2002, 80(17):3201(1-3).



文章编码的识读

为了便于期刊文章的检索、查询、全文信息索取和远程传递以及著作管理,凡具有文献标识码的文章均可标识一个数字化的文章编号,其中 A(理论与应用研究学术论文(包括综述报告))、B(实用性技术成果报告(科技)、理论学习与社会实践总结(社科))、C(业务指导与技术管理性文章(包括领导讲话、特约评论等))三类文章必须编号。

文章编号由期刊的国际标准刊号、出版年、期次号及文章的篇首码和页数五段共 20 位数字组成,其结构为:XXXX-XXXX(YYYY)NN-PPPP-CC,其中 XXXX-XXXX 为文章所在期刊的国际标准刊号(ISSN),YYYY 为文章所在期刊的出版年,NN 为文章所在期刊的期次,PPPP 为文章首页所在期刊页码,CC 为文章页数,“-”为连字符。该编号在全世界范围内是该文章的惟一标识。

文章编号由各编辑部给定,中文文章编号的标识为“文章编号:”或[文章编号]。如:文章编号;1007-2780(2003)02-0097-04 为发表在《液晶与显示》2003 年第 2 期第 97~100 页(共 4 页)上题为“液晶中聚合物网络的形貌研究”(作者:杨文君等)一文的文章编号。