

Article ID: 1007-2780(2010)05-0672-06

Effects of DCJCB Doping Concentration on Performances of Organic Light-Emitting Diodes

LIU Ding-han, ZHANG Fang-hui, MU Qiang
(Department of Electricity and Electronics, Shaanxi University of Science and Technology,
Xi'an 710021, China, E-mail: ldhghan@163.com)

Abstract: A highly efficient white organic light-emitting device could be achieved by changing doping concentration in the emissive layer of the device containing 9,10-di-(2-naph-thyl)anthracene(ADN), 2,5,8,11-tetra-butyl-perylene(TBPe) and 4-(Dicyanomethylene)-2-t-butyl-6-(1,1,7,7-tetramethylj-ulolidyl-9-enyl)-4H-pyran(DCJTB). By examining the device characteristics of uniformly doped devices with varying dopant concentration, it is found that the device with the doping concentration(mass fraction) of 1% DCJTB gains the maximum current(η_L) and power(η_P) efficiencies, which are 6.6 cd/A and 3.21 lm/W at luminance of 10 520 cd/m² with CIE coordinates of (0.318 6, 0.352 0). The different color of devices can be obtained by changing doping concentration of DCJTB.

Key words: OLED; DCJTB; dopant; luminance

DCJTB 掺杂浓度对 OLED 器件性能的影响

刘丁菡, 张方辉, 牟 强
(陕西科技大学 电气与信息工程学院, 陕西 西安 710021, E-mail: ldhghan@163.com)

摘 要: 通过在发光层 ADN:TBPe:DCJTB 中改变 DCJTB 的掺杂浓度, 得到了一种高效的白光 OLED 器件。考察了同一掺杂浓度 TBPe 下不同浓度的 DCJTB 的器件性能, 发现当 DCJTB 掺杂浓度为 1%(质量分数)时, 器件获得最大电流效率 6.6 cd/A 和最大功率效率 3.21 lm/W, 此时亮度为 10 520 cd/m², 对应的 CIE 坐标为 (0.318 6, 0.352 0)。通过改变 DCJTB 浓度, 能够获得不同的器件颜色。

关 键 词: 有机电致发光; DCJTB; 掺杂剂; 亮度

中图分类号: TN383⁺.1 **文献标识码:** A

1 Introduction

Research on white organic light-emitting devices (WOLEDs) has become increasingly important because of their promising applications

in solid state lighting and flat-panel displays^[1]. In particular, the advantages of using white OLEDs are their high efficiency, low operating voltage, and long lifetime^[2-3]. However, WOLEDs for these applications have not been

收稿日期: 2010-02-23; 修订日期: 2010-03-22
基金项目: 浙江省温州市科技计划项目(No. H20080004); 陕西省教育厅专项科研项目(No. 07JK191); 陕西科技大学科研启动基金(No. BJ09-07)
作者简介: 刘丁菡(1984—), 女, 陕西西安人, 硕士, 主要从事有机电致发光显示器件研究。

considered to be realistic because of many technical problems.

White emission from OLEDs can be obtained by mixing three primary colors (red, green, and blue) [4] or two complementary colors (sky blue and orange) [5] in a multi- or a single-layer structure from small molecules or polymers. Generally, WOLEDs based on small molecules have been fabricated in a multilayer structure with one or more emitting layers[6].

Recently, several reports indicated that device performance can be improved substantially by doping with fluorescent or phosphorescent dyes into an active host material in a single emitting layer[7-9]. In this study, we fabricated four groups of OLEDs, which had the same structures except for the emitting layers. We regulated the concentration of red fluorescent dyes DCJTB doping mass fraction from 0~3% in the emissive layer. As a result, the devices with high current and power efficiencies as well as low operating voltages were obtained. Simultaneously, we developed a white organic light-emitting device after doping 2% (mass fraction) of the blue and 1% (mass fraction) of the red dopants. We optimized theoretically and experimentally comprehensive performance of a OLED in charge carrier balance and energy transfer [10]. In addition we observed different enhancement mechanisms, which we attributed to charge trapping phenomena, for devices emitting different colors.

2 Experiments

The schematic structure and the energy-level diagram of OLEDs are shown in Fig. 1. Here, we choose Al/LiF for the cathode. Al/LiF is a good candidate for electrode material because of its superior properties such as high conductivity, high reflectivity in the visible light range, and excellent stability. In addition, Al is a well-known contact material in Si integrated circuits and exhibits good corrosion resistance. We manufactured the OLED using the following

steps. Firstly, 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. Secondly, ITO was cleaned by the standard oxygen plasma treatment for 5 min. Finally, the ITO was handled well in the vacuum chamber.

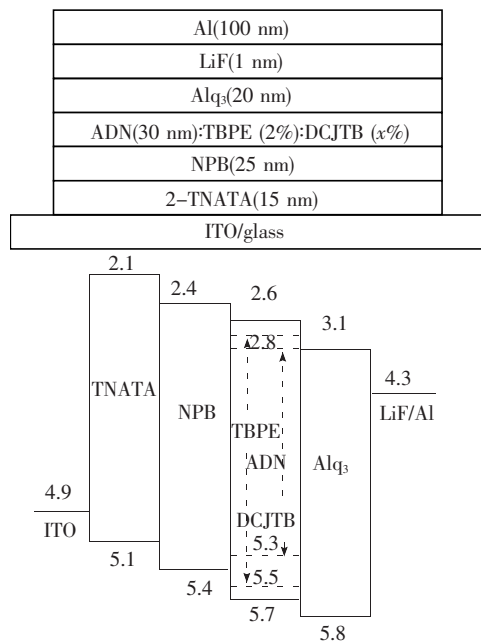


Fig. 1 Schematic structure and energy-level diagram of doped OLED

The organic multilayer devices structured as N-phenyl-amino-triphenylamine(2-TNATA), 4,4-N, N-bis-N-1-naphthyl-N-phenyl-amino-biphenyl(NPB), 2,5,8,11-tetra-butyl-perylene (TBPe) and 4-(Dicyanomethylene)-2-t-butyl-6-(1,1,7,7-tetra-methylj-ulolidyl-9-enyl)-4H-pyran (DCJTB) were doped in a wide band gap blue host 9,10-di-(2-naph-thyl) anthracene (ADN), and tris-(8-hydroxyquinoline) aluminum (Alq₃) consequentially. The concentrations of the guest-to-host molecules were controlled at 2 % (mass fraction) for the TBPe, X% (mass fraction) for the DCJTB (Where X=0,1,2,3). Additionally, the thickness of organic layers were 15 nm for 2-TNATA, and 25 nm, 30 nm, 20 nm for NPB, EML, Alq₃, respectively. All of the materials, including Al/LiF, were deposited by vacuum

evaporation in a vacuum chamber. In a vacuum evaporator, which was evacuated to 6×10^{-4} Pa. The active area of devices was 210 mm^2 . Organic and Al/LiF layers were vacuum-deposited on the ITO surfaces, respectively, deposition rates of 0.2 nm/s for organic and 1 nm/s for Al/LiF, using SI-TM206 six quartz crystal microbalances to control. Four groups of devices were manufactured.

Device A: ITO/2-TNATA (15 nm)/NPB (25 nm)/ADN (30 nm):TBPe (2%):DCJTB (0%)/Alq₃ (20 nm)/ LiF(1 nm)/ Al(100 nm). Device B: ITO/2-TNATA (15 nm)/NPB (25 nm)/ADN(30 nm):TBPe (2%):DCJTB (1%)/ Alq₃ (20 nm)/ LiF(1 nm)/ Al(100 nm). Device C:ITO/2-TNATA(15 nm)/NPB(25 nm)/ADN (30 nm):TBPe (2%):DCJTB (2%)/Alq₃ (20 nm)/LiF(1 nm)/Al(100 nm). Device D:ITO/2-TNATA(15 nm)/NPB(25 nm)/ADN(30 nm):TBPe (2%):DCJTB (3%)/Alq₃ (20 nm)/LiF (1 nm)/Al(100 nm).

Luminance measurement was carried out using a ST-900 spectrophotometer. The EL spectra were taken on a PMS-80 optical multi-channel analyzer with a XYZ stage at room temperature. The current density-voltage characteristics of the device were measured with DC Power Supply PS-3003D measurement unit.

3 Results and Discussion

The J - V and L - V characteristics of the OLED under different doping concentrations are shown in Fig. 2. It can be seen from the four groups of devices that both current density and luminance raise with the increase of driving voltage, and high electric field strength and positive bias voltage is an exponential relationship, showing the typical characteristics of diode rectifier.

We have examined the device characteristics of uniformly doped devices with varying dopant concentration. Under the same TBPe doping level, the current density and luminance of the four

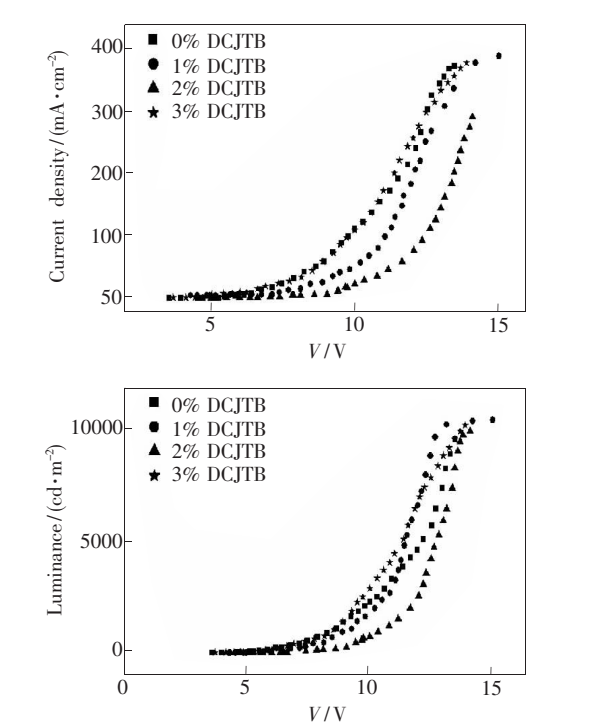


Fig. 2 Current density-voltage-luminance characteristics of the OLED under different doping concentrations

devices show a slight change with variation DCJTB concentration. The maximum luminance is $9\,650\text{ cd/m}^2$ at the breakdown voltage of 13.4 V with none doping concentration of DCJTB in device A. However, the device with the doping concentration of 1% DCJTB gained the maximum luminance of $10\,520\text{ cd/m}^2$ at 15 V, the device B with the doping concentration of 1% gained higher luminance than device A with the none doping concentration. With the doping concentration increasing from 2% to 3%, the corresponding maximum luminance of devices are $10\,020\text{ cd/m}^2$ and $10\,290\text{ cd/m}^2$, respectively.

The current density was low in the device A and it was high in other three devices. The difference in current density in four devices originated from different electron injection from cathode to emitting layer.

Different doping concentration of DCJTB were used to optimize the device performance (Table 1). With the doping concentration of DCJTB increasing from 0 to 1%, the current

(η_{L}) and power (η_{P}) efficiencies increase from 4.2 cd/A to 6.6 cd/A, and 1.85 lm/W to 3.21 lm/W, respectively. From 2% to 3% dopant level(device C~D), the device efficiencies declines. These values collected at 19 mA/cm².

From the results above, the corresponding energy-level diagram was shown in Fig. 1. Carrier injection in OLEDs depends on injection barrier difference from the HTL(or the ETL) and the EML, the highest occupied molecular orbital (HOMO) level and the lowest unoccupied molecular orbital(LUMO) level of ADN are 5.7 eV and 2.6 eV, respectively. The HOMO and LUMO of TBPe dopant are 5.5 eV and 2.8 eV, respectively. The energy gap of HOMO between NPB and TBPe is 0.3 eV, while the LUMO between TBPe and Alq₃ is 0.5 eV. Therefore, hole and electron carriers have encountered energy

barrier for injection to the EML. However, the HOMO and LUMO of DCJTB dopant are 5.3 eV and 3.1 eV, respectively. There is no energy barrier for hole injection between the hole transport layer to the emitting layer. Simultaneously, there is only 0 for electron injection between the electron transport layer to the emitting layer. Electrons are directly injected from Alq₃ layer to ADN doped DCJTB and TBPe emitting layer without any energy barrier. This structure forms highly energy transfer, which leads to a high current density and efficiencies. Oppositely, highly doped (3%) device exhibites relatively low current and power efficiencies than that of device B. This is attributed to the exciton quenching by the accumulated holes and electrons^[11-13], and electric field induced dissociation of excitons^[13], which occur in the heavily doped EML.

Table 1 Electroluminescence performance of OLEDs A~D

Device	Dopant(%)		$V_{\text{turn-on}}$ (V)	Luminance (cd/m ²)	$\eta_{\text{L}}^{\text{c}}$ (cd/A)	$\eta_{\text{P}}^{\text{c}}$ (lm/W)	CIE ^c
	DCJTB	TBPe					
A	0	2	3.5	1 384 ^a 9 650(13.4) ^b	4.2	1.85	(0.148 8, 0.201 6)
B	1	2	4.2	2 010 ^a 10 520(15) ^b	6.6	3.21	(0.318 6, 0.352 0)
C	2	2	4.7	2 020 ^a 10 020(14) ^b	6.4	2.48	(0.424 8, 0.415 1)
D	3	2	3.6	1 400 ^a 10 290(13.8) ^b	4.5	2.18	(0.148 , 0.201 6)

^a values collected at 30 mA/cm².

^b maximum values. values of the maximum luminance under the breakdown voltage

^c At 19 mA/cm²

Fig. 3 shows the EL spectra of the OLEDs with different doping concentration and the corresponding CIE coordinates in the chromaticity diagram. Judging from the EL intensity versus wavelength characteristics, it can be seen that the different doping concentration of DCJTB has great influence on the color of devices. Electroluminescence spectra of four devices showed dif-

ferent maximum luminescence wavelength. The no doping device shown a pure blue OLED with CIE(*x*, *y*)color coordinates of (0.148 8, 0.201 6), which gives one emission peak at 461 nm (with a shoulder at 491 nm). While the device with the doping concentration of 1% DCJTB gains a stable white OLED with CIE(*x*, *y*) color coordinates of (0.318 6, 0.352 0), which emits two

peaks at 464 nm (with a shoulder at 492 nm) and 565 nm. The device with the doping concentration of 2% (mass fraction) DCJTB shows a slight yellow OLED with CIE(x , y) color coordinates of (0.424 8, 0.415 1), which also gives two emission peaks at 461 nm (with a shoulder at 491 nm) and 575 nm. The last device with the doping concentration of 3% DCJTB gains a orange OLED with CIE(x , y) color coordinates of (0.525 9, 0.440 2), which has only one peak at 589 nm. From these results, we speculate that carrier recombination of electrons and holes mainly occur in the EML after doping DCJTB.

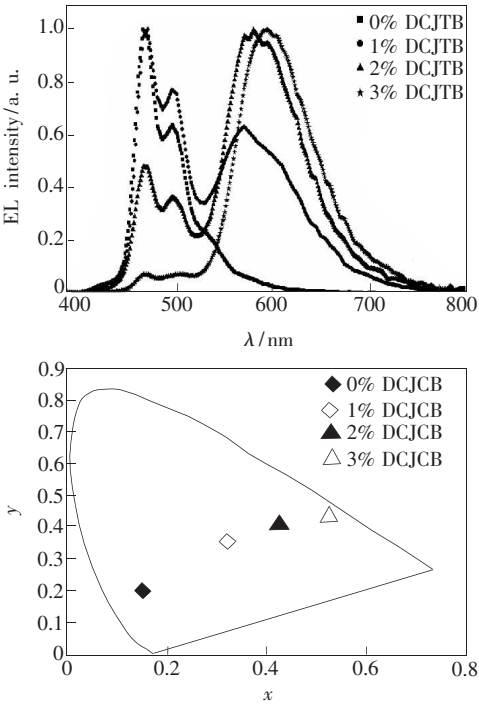


Fig. 3 EL spectra of OLEDs with different doping concentration and the corresponding CIE coordinates in the chromaticity diagram.

Fig. 4 displays the EL spectra of a stable WOLED with 1% DCJTB gives two emission peaks at 462 nm(with a shoulder at 492 nm) and 565 nm. Unlike common WOLED, which typically suffers from a color change with driving conditions, there is practically no EL color shift with various driving currents in our device, suggesting

a balanced carrier injection in this device structure. For instance, the CIE color coordinates of white OLED change only from (0.321 7, 0.356 5), (0.318 6, 0.352 0), (0.315 6, 0.347 7) and (0.313 2, 0.348 8) to (0.312 3, 0.343 7) as the current density increases gradually from 9.5 mA/cm², 19 mA/cm², 28.5 mA/cm², and 38.1 mA/cm² to 47.6 mA/cm², respectively. Such device structure provides an improved color stability as the aging rate is determined by only one emitters^[14].

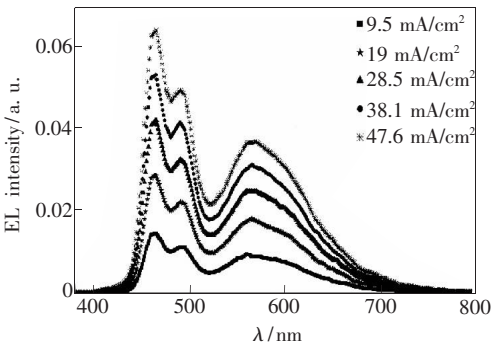


Fig. 4 EL spectra of WOLED at varying current densities

4 Conclusion

In summary, We present the effects of DCJTB on emitter ADN:TBPe:DCJTB in this paper. The results show that different doping concentration of DCJTB has great influence on the color of devices, 1% DCJTB in ADN:TBPe:DCJTB can realize stable and pure white emission with CIE coordinates of (0.318 6, 0.352 0), and 0%, 2%, 3% DCJTB for blue, yellow, orange emission respectively. Simultaneously, devices luminance and efficiency can be optimized by DCJTB ratio, device obtain higher efficiency and maximum brightness with assist dopant TBPe, 1% DCJTB devices have a maximum brightness of 10 520 cd/m² and luminance of 6.6 cd/A. By analyzing the impact of DCJTB on device performance, we have learned more in-depth practical and theoretical understanding of the energy transfer and the carrier capture mechanism.

References:

- [1] Tang C W, VanSlyke S A. Organic electroluminescent diodes [J]. *Appl. Phys. Lett.*, 1987, 51(12):913-915.
- [2] Matsushima T, Adachi C. Extremely low voltage organic light-emitting diodes with p-doped alpha-sexithiophene hole transport and n-doped phenyldipyrrenylphosphine oxide electron transport layers [J]. *Appl. Phys. Lett.*, 2006, 89(25):253506(1-3).
- [3] Meerheim R, Walzer K, Pfeiffer M, *et al.* Ultrastable and efficient red organic light emitting diodes with doped transport layers [J]. *Appl. Phys. Lett.*, 2006, 89(6): 061111(1-3).
- [4] Lee YG, Kee I S, Shim H S, *et al.* White organic light-emitting devices with mixed interfaces between light emitting layers [J]. *Appl. Phys. Lett.*, 2007, 90(24):243508(1-3).
- [5] Schwartz G, Fehse K, Pfeiffer M, *et al.* Highly efficient white organic light emitting diodes comprising an interlayer to separate fluorescent and phosphorescent regions [J]. *Appl. Phys. Lett.*, 2006, 89(8): 083509(1-3).
- [6] Lee T W, Park O O, Cho H N, *et al.* White emission from a ternary polymers blend by incomplete cascade energy transfer [J]. *Synth. Met.*, 2001, 122:437-441.
- [7] Niu Y H, Ma H, Xu Q, *et al.* High-efficiency light-emitting diodes using neutral surfactants and aluminum cathode [J]. *Appl. Phys. Lett.*, 2005, 86(8):083504(1-3).
- [8] Guo T F, Yang F S, Tsai Z J, *et al.* Organic oxide/Al composite cathode in efficient polymer light-emitting diodes [J]. *Appl. Phys. Lett.*, 2006, 88(11):113501(1-3).
- [9] Guo T F, Yang F S, Tsai Z J, *et al.* Organic oxide/Al composite cathode in small molecular organic light-emitting diodes [J]. *Appl. Phys. Lett.*, 2006, 89(5):053507(1-3).
- [10] Wang Q, Deng Z Q, Ma D G. Realization of high efficiency microcavity top-emitting organic light-emitting diodes with highly saturated colors and negligible angular dependence [J]. *Appl. Phys. Lett.*, 2009, 94(23):233306(1-3).
- [11] Adachi C, Baldo M A, Forrest S R. Electroluminescence mechanisms in organic light emitting devices employing a europium chelate doped in a wide energy gap bipolar conducting host [J]. *J. Appl. Phys.*, 2000, 87(11): 8049-8055.
- [12] Baldo M A, Adachi C, Forrest S R. Transient analysis of organic electrophosphorescence. II. Transient analysis of triplet-triplet annihilation [J]. *Phys. Rev. B*, 2000, 62(16):10967-10977.
- [13] Reineke S, Walzer K, Leo K. Triplet-exciton quenching in organic phosphorescent light-emitting diodes with Ir-based emitters [J]. *Phys. Rev. B*, 2007, 75(12):125328-125340.
- [14] Ho C L, Lin M F, Wong W Y. High-efficiency and color-stable white organic light-emitting devices based on sky blue electrofluorescence and orange electrophosphorescence [J]. *Appl. Phys. Lett.*, 2008, 92(8):083301(1-3).