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Power Efficiency Improvement of White Phosphorescent Organic Light-Emitting Diode with Thin Double-Emitting Layers and Hole-Trapping Mechanism

Fuh-Shyang Juang*, Lin-Ann Hong, Shun-Hsi Wang, Yu-Sheng Tsai,
Ming-Hong Gao, Yun Chi[1], Han-Ping Shieh[2], and Jen-Sung Hsu[3]

Institute of Electro-Optical and Materials Science, National Formosa University, Yunlin 632, Taiwan
[1] *Department of Chemistry, National Tsing Hua University, Hsinchu 300, Taiwan*
[2] *Department of Photonics and Display Institute, National Chiao Tung University, Hsinchu 300, Taiwan*
[3] *Chemistry Department, Chung-Shan Institute of Science and Technology, Lungtan 325, Taiwan*

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This study is carried out to discuss how to reduce the driving voltage of blue phosphorescent organic light-emitting diodes (PHOLEDs) by using a thin double-emission layer. A hole transport-type host (TCTA) is inserted between the hole transport layer (TAPC) and the emitting layer (EML), constituting a buffer layer between them with the aim of improving charge carrier balance. Furthermore, in this study, we also utilize the interface between double light-emitting layers of devices by codoping them with a red phosphorescent dopant [$Os(bpftz)_2(PPh_2Me)_2$]. An Os complex with a high-lying highest occupied molecular orbital (HOMO) energy level (trapping holes) is codoped at the interface between emitting layers and an exciton-formation zone is expanded to obtain a white PHOLED with high efficiency. From the results, the optimal structure of the white device exhibits a yield of 35 cd A^{-1}, a power efficiency of 22 lm W^{-1}, and CIE coordinates of (0.33, 0.38) at a luminance of 1000 cd m^{-2}. Furthermore, the power efficiency can be improved to 30 lm W^{-1} by attaching the outcoupling enhancement film. © 2011 The Japan Society of Applied Physics

1. Introduction

Recently, structures that can improve the efficiency of organic light-emitting diodes (OLEDs) have been continuously reported, e.g., one emitting layer is inserted at the heterogeneous interface of devices to increase luminance efficiency.[1)] Other reports also validate that hole-buffer layers can be used to effectively help hole injection, reduce the driving voltage of devices, and improve the power efficiency.[2)] Additionally, compared with a single-emitting layer, double-emitting layers can effectively extend the exciton-formation zone and reduce the efficiency roll-off.[3–5)] Because the triplet–triplet annihilation is the main mechanism for the efficiency decrease of a phosphorescent organic light-emitting diode (PHOLED) at high luminance.[5,6)] In addition, Leo *et al.*, Baldo *et al.*, and Kepler *et al.* reported that a weak exciton-formation zone and a large triplet exciton density inside the EML will cause the quantum efficiency decay quickly (roll-off).[6–8)] Therefore, using a thin double-EML structure and controlling the recombination zone (RZ) at the interface of the two EMLs can extend the exciton-formation zone to decrease the carrier accumulation[9)] and reduce the driving voltage. To further improve the charge carrier balance, in this study, we inserted a hole transport-type host (TCTA) between the hole transport layer (HTL) and the EML[10)] to adjust the hole injection current and further increase device efficiency. Moreover, the orange-red-emitting phosphor [$Os(fptz)_2(PPh_2Me)_2$] was doped at the interface of the two EMLs. By using the ability of hole trapping in $Os(fptz)_2(PPh_2Me)_2$, the holes were trapped at the interface of the double-emission layers, avoiding the color shift of the white OLED device.

2. Experimental Methods

White PHOLEDs were fabricated on prepatterned ITO substrates with a sheet resistance of $13.59 \pm 5\,\Omega\,\mathrm{square}^{-1}$. The substrates were cleaned using acetone, isopropyl alcohol, and DI water followed by O_2 plasma treatment. All the organic and metal layers were vacuum-deposited at 2×10^{-6} Torr. 1,1-Bis[(di-4-tolylamino)phenyl]cyclohexane (TAPC) was used as a hole transport layer, 4,4′,4″-tris(N-carbazolyl)triphenylamine (TCTA)[3)] was used as a hole buffer layer and a hole transport-type host, 2,6-bis[(3-carbazol-9-yl)phenyl)]pyridine (26DCzPPy) was used as a bipolar host, iridium-bis[(4,6-(difluorophenyl-pyridinato-N,C2)] (FIrpic) was used as a blue dopant, doped with 10 wt % of $Os(bpftz)_2(PPh2Me)_2$[11,12)] as the red dopant, and tris[3-(3-pyridyl)-mesityl]borane (3TPYMB)[13)] was used as the ETL. The cathode consisted of 0.5-nm-thick LiF or CsF followed by 200-nm-thick Al. Finally, a brightness enhancement film (BEF) is attached onto the glass substrate to further improve the performance. Figure 1 shows the energy band structure of the white device with $Os(bpftz)_2$-$(PPh2Me)_2$ doped at the double-EML interface. Spectra Scan PR650 and Keithley 2400 were employed to measure the current–voltage–luminance (I–V–L) characteristics.

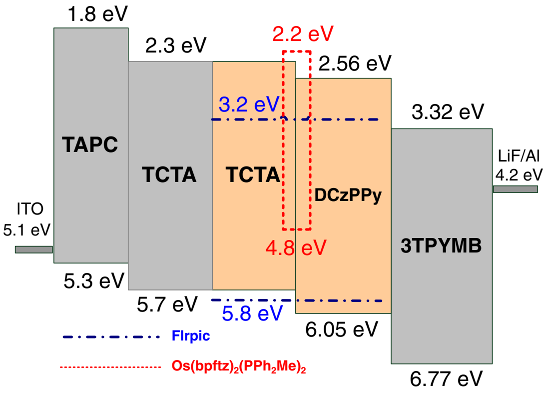

Fig. 1. (Color online) Energy band structure of white PHOLED.

3. Results and Discussion

The single-emitting-layer (single-EML) blue PHOLED used in this study is made of ITO/TAPC (20 nm)/TCTA (X nm)/17 wt % FIrpic:26DCzPPy (20 nm)/3TPYMB (35 nm)/LiF/Al. Figure 2 shows the normalized electroluminescence (EL) spectra of blue PHOLEDs with different buffer layer

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*E-mail address: fsjuang@seed.net.tw

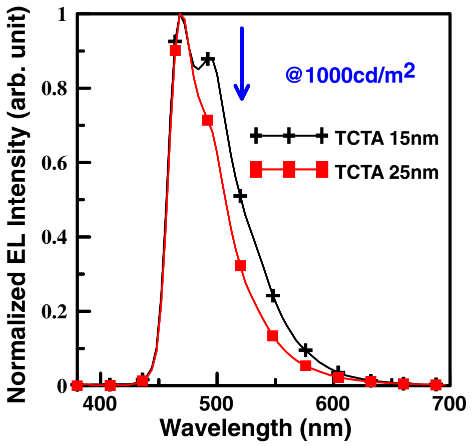
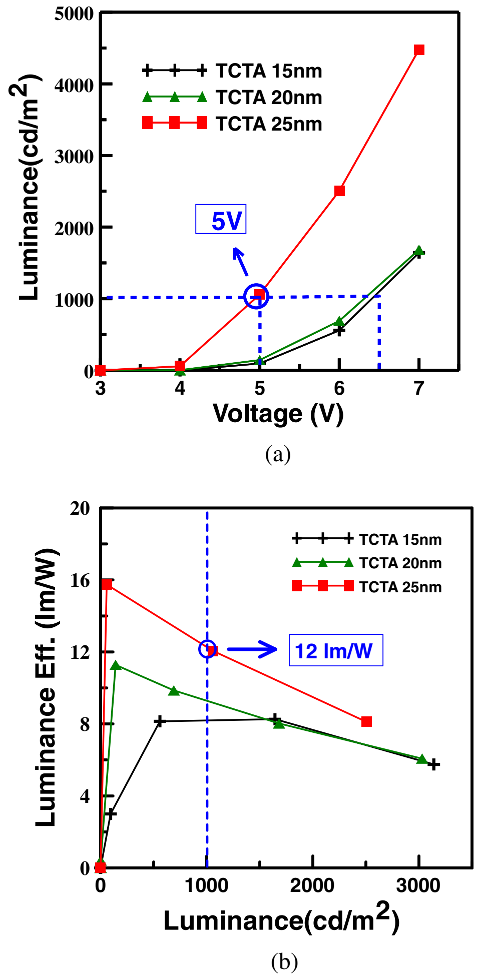


Fig. 2. (Color online) EL spectra of blue PHOLEDs with different interlayer (TCTA) thicknesses.

thicknesses. The results suggest that the RZ of the device with a 15 nm interlayer (TCTA) is positioned at the center of the EML.[14,15] However, the charge carrier is unbalanced in most OLED devices owing to the large difference between hole and electron mobilities. Therefore, to further improve the driving voltage and efficiency, the thickness of the interlayer (TCTA) was adjusted (from 10 to 25 nm). From Fig. 2, it can be observe that the shoulder peak at approximately 500 nm decreased in intensity when the thickness of the interlayer increased (TCTA 25 nm). Lee and coworkers have reported that the change in emission intensity at approximately 500 nm was attributed to the optical effect caused by the RZ shift.[14,15] Therefore, it can be suggested that the hole injection current increased when the interlayer (TCTA) thickness increased. Moreover, the RZ of devices can be controlled near the EML/ETL interface, which resulted in the improvement of the recombination of charge carriers. From the results, the blue device showed that the driving voltage decreased to 5 V and the power efficiency increased to 12 lm W^{-1} at the luminance of 1000 cd m^{-2} [as shown in Figs. 3(a) and 3(b)]. The power efficiency was rolled off when the luminance increased. It has also been reported that triplet–triplet annihilation is proportional to the square of the triplet exciton density, and a narrow emission zone has been found to have a negative effect on triplet–triplet annihilation owing to its high triplet exciton density.[16)] In the single blue-emitting OLED device, therefore, a drop as high as 63% in efficiency roll-off was observed from 1 to 3000 cd m^{-2}. Moreover, we used CsF as an electron injection layer (EIL) to increase the electron injection current owing to the V_{BI} of CsF/Al (2.55 ± 0.05) being higher than that of LiF/Al (2.31 ± 0.05).[18)] From Fig. 4, we can observe that the power efficiency of the blue device with CsF was increased to 17 lm W^{-1}. Next, we used a double-EML structure in the blue device to reduce the thickness of the EML, which will reduce the driving voltage, improve the charge carrier injection, and extend the exciton-formation zone. The device structure is ITO/TAPC (50 nm)/12 wt % FIrpic:TCTA (5 nm)/17 wt % FIrpic:26DCzPPy (5 nm)/3TPYMB (50 nm)/CsF/Al. In this study, because there is an excessively large gap in hole mobility between the hole transmission layer (TAPC) ($\mu_h \sim 1.0 \times 10^{-2}$ cm^{-2} V^{-1} s^{-1}) and the electrical transmission layer (3TPYMB) ($\mu_h \sim \times 10^{-5}$ cm^{-2} V^{-1} s^{-1}), leading to extreme unbalance in carriers, we insert a hole buffer layer between the HTL (TAPC) and the EML. The hole buffer layer ws substituted for part of TAPC. The device structure is ITO/TAPC (40 nm)/TCTA (10 nm)/



(a)

(b)

Fig. 3. (Color online) (a) Luminance–voltage and (b) power efficiency–luminance characteristics of blue PHOLEDs with different interlayer (TCTA) thicknesses.

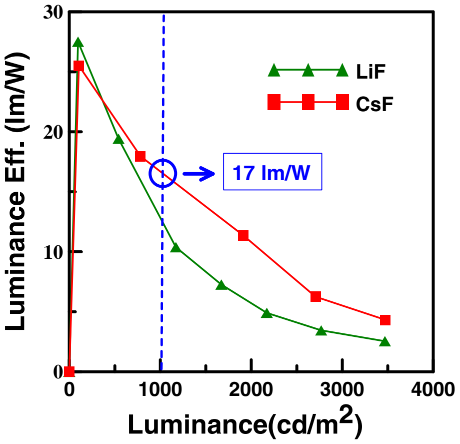


Fig. 4. (Color online) Power efficiency–luminance characteristics of blue PHOLEDs with different EIL materials.

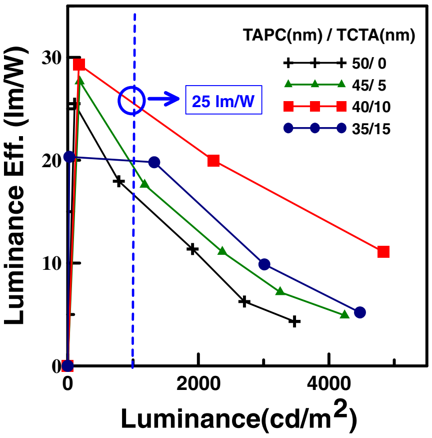


Fig. 5. (Color online) Power efficiency–luminance characteristics of blue PHOLEDs with different buffer layer (TCTA) thicknesses.

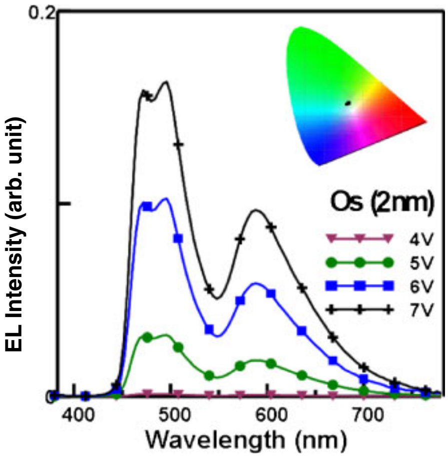


Fig. 6. (Color online) Electroluminescence (EL) spectra of white device at different driving voltages.

Table I. Optoelectronic properties of different buffer layer (TCTA) thicknesses at 1000 cd m^{-2}.

TCPC (nm)	TCTA (nm)	V (V)	I (mA m^{-2})	Y (cd A^{-1})	P (lm W^{-1})
50	0	5.2	4.5	27	17
45	5	4.8	4.0	30	19
40	10	4.4	3.2	35	25
35	15	4.7	3.2	30	20

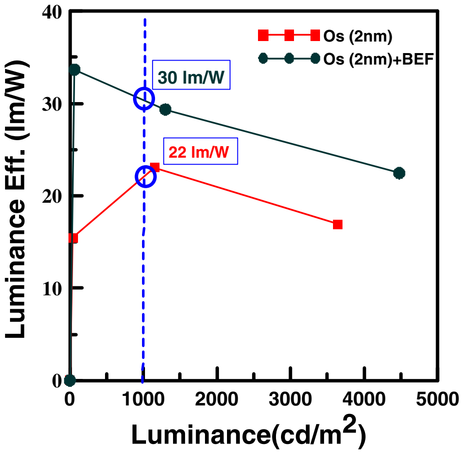


Fig. 7. (Color online) Power efficiency–luminance characteristics of white PHOLEDs.

12 wt % FIrpic:TCTA (5 nm)/17 wt % FIrpic:26DCzPPy (5 nm)/3TPYMB (50 nm)/CsF/Al. It can be seen from the power efficiency–luminance curves in Fig. 5 and Table I that after the insertion of a hole buffer layer (TCTA), the current density of the device with (TAPC/TCTA = 40/10) is less than that of the device with (TAPC/TCTA = 50/0) without the inserted buffer layer at a luminance of 1000 cd m^{-2}; thus, it increases the luminance efficiency of devices (see Table I). In the blue double-emitting OLED device, a 41% drop in efficiency roll-off was noted from 1 to 3000 cd m^{-2}, because the hole mobility of the TCTA buffer layer ($\mu_h \sim 10^{-4}$ cm^{-2} V^{-1} s^{-1}) is less than that of the hole transmission material (TAPC), it adjust a hole injection current and further increase device efficiency. Experimental results show that when TCTA (10 nm) is used as the buffer layer, the luminance efficiency of the blue device increases up to 35 cd A^{-1} at a luminance of 1000 cd m^{-2}, the power efficiency reaches 25 lm W^{-1}, and the driving voltage decreases to 4.4 V (as shown in Table I).

Furthermore, this study also utilizes double emitting layers (D-EMLs) to enlarge the exciton-formation zone of devices for reducing the thickness of the EML and the driving voltage for blue PHOLEDs. In the experiment, TCTA and 26DCzPPY are used as the host materials of D-EMLs; FIrpic is a well-known electron transport-type blue dopant, TCTA is a hole transport-type host material, and 26DCzPPY is a bipolar transport-type host material. Therefore, by utilizing the difference in the transport capability of the materials in the D-EMLs, in combination with the doping concentration of FIrpic, the recombination zone can be controlled to be at the interface between TCTA and 26DCzPPY.5) Finally, Os(bpftz)$_2$(PPh2Me)$_2$ is doped at the interface between two blue EMLs, and the thicknesses of these layers are adjusted to be optimal; thus, a white device with high efficiency will be obtained with a device structure of ITO/TAPC (40 nm)/TCTA (10 nm)/12 wt % FIrpic:TCTA (4 nm)/2 wt % Os:12 wt % FIrpic:TCTA (1 nm)/2 wt % Os:17 wt % FIrpic:26DCzPPy (1 nm)/17 wt % FIrpic:26DCzPPy (4 nm)/3TPYMB (50 nm)/CsF/Al. Figure 1 shows the energy band structure of the optimal white device. Because Os can trap holes (with a high-lying HOMO level) at the interface between two EMLs, the recombination zone of devices will be confined at the interface between two EMLs. It does not only enlarge the exciton-formation zone and increase the device efficiency, but also reduce the color shift caused by driving voltage changes of devices. However, as can be seen from the electroluminescence (EL) spectra of the white device at different voltages in Fig. 6, the intensity of the red wavelength gradually decreases, which is caused by the decrease in energy transfer rate from FIrpic to Os when the driving current density increases,17) leading to blue shift. From the results of the above experiments shown in Fig. 7, the luminance efficiency of the white device is expected to reach 35 cd A^{-1} at a luminance of 1000 cd m^{-2}. In the double-emitting WOLED device, a 23% drop in efficiency roll-off was observed from 1 to 3000 cd m^{-2}. The power efficiency is expected to reach 22 lm W^{-1}, the CIE coordinates are expected to be (0.33, 0.38), and the driving voltage is expected to decrease to 4.9 V. Furthermore, the efficiencies can be increased to 35 cd A^{-1} and 30 lm W^{-1} (with a driving voltage of 4 V) by attaching a brightness enhancement film (BEF) onto the glass substrate.

4. Conclusions

By inserting a hole transport-type host (TCTA) as a buffer layer in this study, the quantity of holes injected into the EML is effectively controlled, and the balance of charge carriers is realized. Then, by expanding the recombination zone of devices by using structures of double EMLs, a red phosphorescent dopant [$Os(bpftz)_2(PPh2Me)_2$] featuring hole traps is codoped at the interface (FIrpic:TCTA/FIrpic:DCzPPy), and by adjusting the thickness of each organic layer to be optimal, a white PHOLED with high efficiency is obtained. When the luminance of white devices is 1000 cd m^{-2}, the driving voltage decreases to 4.9 V, the luminance efficiency reaches 35 cd A^{-1}, and the power efficiency increases up to 22 lm W^{-1}. Furthermore, the efficiencies can be increased to 35 cd A^{-1} and 30 lm W^{-1} (with a driving voltage of 4 V) by attaching a brightness enhancement film.

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