

## High Efficiency Very Low Color Temperature Phosphorescent Organic Light Emitting Diodes

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**Abstract.** Color temperature of light plays an important role in human physiology and psychology as low color temperature illumination source shows mild effect on melatonin suppression. We presents an energy efficient very low color temperature (CT) organic light emitting diodes using a phosphorescent orange emitter tris(2-phenylquinoline) iridium(III). The device exhibits a high power efficiency of 44.3 and ~32 lm/W at 100 and 1,000 cd/m<sup>2</sup>, respectively. The device also reveals a very low color temperature, ~1,745 K, with 19.2% external quantum efficiency and (0.571, 0.424) Commission International de L'Eclairage (CIE) coordinates at 1,000 cd/m<sup>2</sup>. The high efficiency may be attributed to good energy transfer between host and guest materials, and balanced carrier injection.

**Keywords:** High efficiency, low color temperature, physiologically/psychologically friendly, light at night.

### 1. Introduction

Since the first report of OLEDs by Tang and Van-Slyke in 1987, numerous approaches have been used to improve the device efficacy.<sup>[1]</sup> To achieve the highest efficiencies for phosphorescent OLEDs, both carriers and excitons must be well confined within a emission layer.<sup>[2-5]</sup> Nowadays, OLEDs are widely recognized as a potential application for high quality lightings.<sup>[6]</sup> Color temperature of light also plays a crucial role in human physiology and psychology.<sup>[7-13]</sup> Since high CT light stimulates the secretion of cortisol, a hormone that keeps people awake and energetic.<sup>[7,9,13]</sup> While, a high CT light remarkably suppresses the nocturnal secretion of oncostatic melatonin (MLT), increasing the risk of malignancy in breast, colorectal and prostate.<sup>[10]</sup> While the low CT light source has a slighter effect on MLT suppression and provide the warm feeling.<sup>[11-13]</sup> Hence, developing a highly energy efficient very low CT lighting is an important task, particularly a potential lighting source at night.

A few conventional and mainstream lighting devices (candles, incandescent bulbs and warm-white fluorescent lamps) have relatively low CT, e.g., candles have a CT ~2,000 K, while the incandescent bulbs have a CT 2,000 to 2,500 K, depending on wattage. However, both candles and incandescent bulbs are energy wasting illumination sources. OLEDs have provided a novel and efficient alternative for solid-state lighting, which is energy saving and CT tailorable.<sup>[14,15]</sup> Nowadays, numerous research groups are working to achieve a considerably high efficiency for commercial accessibility of OLEDs. Leo's group reported a power efficiency 38 lm/W with 3,180 K,<sup>[16]</sup> So et al. achieved 40lm/W with 4,970 K,<sup>[17]</sup> Kido and his co-workers reported 55 lm/W efficacy with 5,340 K,<sup>[18]</sup> Forrest et al. achieved 12 lm/W with 4,750 K.<sup>[19]</sup> While, Jou's group reported a very low CT (1,770K) and efficacy 26 lm/W at 1,000 cd/m<sup>2</sup>.<sup>[20]</sup> Recently, their group have reported the efficacy ~29 lm/W with 1,940 K.<sup>[21]</sup> As well known that the lowest CT of daylight is 2,500 K. To attain a CT lower than 2,500 K, long wavelength emitters are requisite.

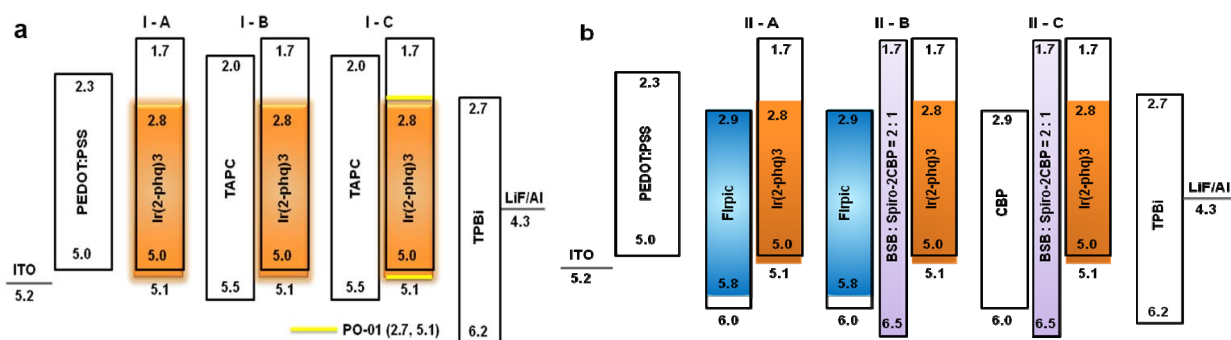
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This study demonstrates both single and double emission layer (EML) based highly efficient very low CT OLEDs with and without electron confining layer (ECL) and various carrier modulation layers (CML), respectively. Single EML based device architecture showed the efficacy  $\sim 32$  lm/W ( $\sim 43$  cd/A) with 1743K at 1000 cd/m<sup>2</sup>. While the best double EML based device revealed the efficacy  $\sim 29$  lm/W (49.5 cd/A) with 1,940 K at 1,000 cd/m<sup>2</sup>. The CTs of single layer devices are ranging from 1,743 to 1,965 K and for double layer 1,770 to 1,940 K. Device I-B exhibits the highest efficacy with an ultimate low CT in all proposed devices. Thus, the proposed Device I-B may be an ideal lighting source for light at night to safeguard human health.

## 2. Experimental

As revealed in Fig. 1(a), Single EML device composed of a 125 nm indium tin oxide (ITO) anode layer, a 25 nm poly(3,4-ethylene-dioxythiophene)-poly-(styrene-sulfonate) (PEDOT:PSS) hole transporting layer (HTL), a 15 nm di-[4-(N,N-ditoly-amino)-phenyl]cyclohexane (TAPC) electron confining layer (ECL), a 20 nm orange red EML, a 32 nm 1,3,5-tris(N-phenyl-benzimidazol-2-yl)benzene (TPBi) electron transporting layer (ETL), a 1 nm lithium fluoride (LiF) and a 150 nm aluminum (Al) cathode layer. The EML is composed of the 4 wt% of the orange-red dye tris(2-phenylquinoline) iridium(III) [Ir(2-phq)<sub>3</sub>] doped in a low hole injection based host 2,7-bis(carbazol-9-yl)-9,9-ditolyfluorene (Spiro-2CBP). Whilst the double EML based device (Fig. 1(b)) composition is 125 nm (ITO)/35 nm (PEDOT:PSS)/10nm blue EML [(4,40-bis(carbazol-9-yl) biphenyl (CBP) doped with (16 wt% bis(4,6-difluorophenylpyridinato-N, C2)picolinato-iridium(III) (FIrpic)]/0 or 3 nm blend interlayer (BSB: Spiro-2CBP)/5 nm orange red EML (4wt % Ir(2-phq)<sub>3</sub> doped with Spiro-2CBP)/32 nm TPBi/1nm LiF/150 nm Al.



**Fig. 1:** The energy diagram of (a) the studied single EML devices using spiro-2CBP host and Ir(2-phq)<sub>3</sub> dye with and without ECL (TAPC) and Device I-C had a 5 wt% PO-01 co-host and (b) Double emissive layer devices using CBP host for blue EML and Spiro-2 CBP for orange EML, Device II-B to II-D all had a 3 nm blend interlayer (BSB: Spiro-2CBP).

The EL characteristics and CIE of the resultant low CT OLEDs were measured by using a Minolta CS-100 luminance-meter and a PR-655 Spectra Scan-spectrophotometer. Current-voltage (I-V) characteristics were measured with a Keithley 2400 electrometer.

## 3. Result and discussion

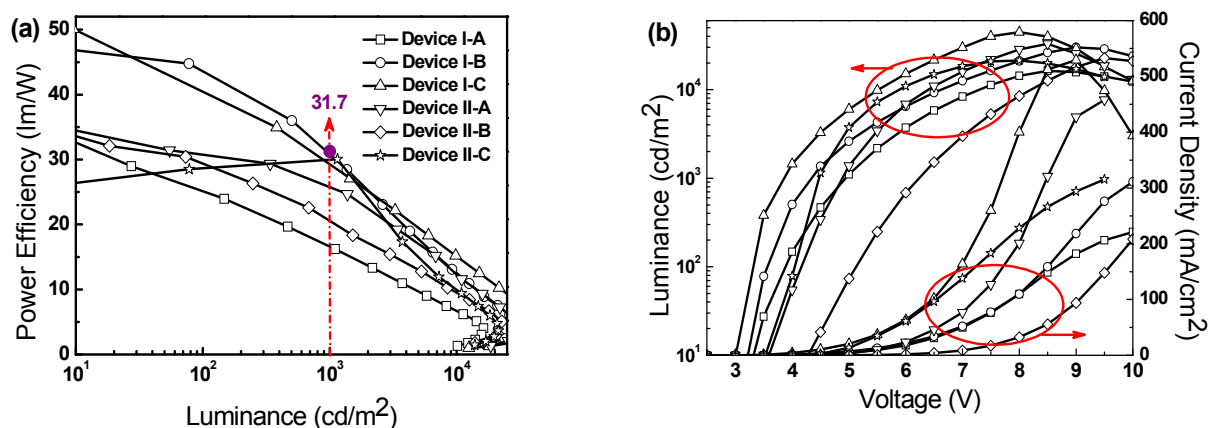
Table I compiles the electroluminescent properties of the studied single and double EML devices. The current efficiency of a single EML based device without an ECL (Device I-A) is 26.2 cd/A with a CT of 1,965 at 1,000 cd/m<sup>2</sup>. The current efficiency markedly increases by 64% (42.9 cd/A) with 1,743 K upon adding a 15 nm TCTA electron confining layer (Device I-B). At the same time power efficiency and external quantum efficiency improves from 16.8 to 31.7 lm/W ( $\sim 89\%$ ) and 10.8 to 19.2% at 1,000 cd/m<sup>2</sup>, respectively. This significant improvement may be attributed to the fact that the TAPC possesses a hole-modulation function.<sup>[15]</sup> However, the current efficiency abruptly decreases by 18% (36.4 cd/A) upon adding a 5 wt% PO-01 as co-host. A double EML based Device II-B reveals the maximum current efficiency 49.5 cd/A (29.1 lm/W and 20.8%) with a CT of 1,940 K.

**Table I:** EL characteristics of the both single emission layer and double emission layer devices, comparing with and without co-host based device and effect of CML and blue EML composition on OLEDs performance

For single emission layer device architecture								
Device	ECL	Co-host	Driving voltage at 10 cd/m <sup>2</sup> (V)	EQE	$\eta_p$	$\eta_c$	CT	1931 CIE color-coordinate at 1000 cd/m <sup>2</sup>
				(%)	(lm/W)	(cd/A)	(K)	
I-A	--	--	3.2	12.8/10.8	25.9/16.8	31.2/26.2	1962/1965	(0.55, 0.44)
I-B	TAPC	--	3.1	22.0/19.2	44.3/31.7	49.7/42.9	1743/1743	(0.57, 0.43)
I-C	TAPC	PO-01	3.0	20.2/16.0	46.4/30.4	45.7/36.4	1778/1793	(0.57, 0.43)
For double emission layer device architecture								
Device	CML	Blue EML	Driving voltage at 10 nits (V)	EQE (%)	$\eta_p$ (lm/W)	$\eta_c$ (cd/A)	CT (K)	1931 CIE color-coordinate at 1000 cd/m <sup>2</sup>
	BSB/Spiro-2CBP	CBP/Flrpic						
	II-A	--	1/0.16	3.6	17.4/17.2	30.8/25.6	40.6/39.9	1840/1860
II-B	2/1	1/0.16	4.1	22.7/20.8	36.0/29.1	53.8/49.5	1880/1940	(0.55, 0.43)
II-C	2/1	1/-	3.5	17.2/18.8	28.6/30.0	36.2/39.8	1760/1770	(0.57, 0.43)

*ECL*-Electron confining layer, *EQE*-External quantum efficiency,  $\eta_p$ -Power efficiency,  $\eta_c$ -Current efficiency, *CT*-Color temperature

Fig. 1(a) shows the energy diagram of the all three single EML devices. These single layer devices possess excellent hole-confinement properties, but its electron confinement is rather poor. To improve the electron confinement, a thin layer of TAPC is incorporated in the device. As also shown in Fig. 1 (a), the HOMO level of TAPC is 0.4eV lower than of PEDOT:PSS, which may balance the carrier injection and improve the device performance.<sup>[22]</sup> Besides better carrier injection the high efficiency may be attributed to its emissive layer architecture. The single EML based Device I-B can effectively prevent carriers being trapped in Ir(2-phq)<sub>3</sub> and leading excitons to form on the host rather than the guest, which would in turn results in high device efficiency. The power efficiency of single EML based device approximately doubled, at 1,000 cd/m<sup>2</sup>, upon inclusion of 15 nm ECL (TAPC). While, the device power efficiency unexpectedly drops upon adding the PO-01 as co-host, it may be attributed due to the insufficient energy transfer from spiro-2CBP to co-host (PO-01) emitter.(see Fig. 2a)



**Fig. 2:** (a) Power efficiency versus luminance and (b) current density and luminance versus applied bias, for both single and double EML devices, as shown in Fig. 2a, the PE of Device I-B and II-B are higher than those of other devices at various luminances, but Device II-C unexpectedly shown the highest value of PE at 1,000 cd/m<sup>2</sup>. This may be because Device II-C has the lowest applied voltage, even though its EQE and CE were lower than those of Device II-B

Fig. 1(b) shows the energy diagram of double EML low CT- OLEDs. The efficiency of the device without a carrier modulating layer (CML) is markedly enhanced, upon incorporation of BSB and Spiro-2CBP blend interlayer (weight ratio-2:1) between blue and orange red EMLs. Comparing Device II-A with Device II-B, the EQE (power efficiency and current efficiency) increased from 17.2% to 20.8% (25.6 to 29.1 lm/W and

39.9 to 49.5 cd/A) with 1,940K. The EQE of Device II-B exceeded the 20% theoretical limit, suggesting this device has near perfect carrier-injection balance.

In Device II-C, the blue emitter was removed from the first emissive layer and only CBP host was deposited instead of first emissive layer. Device shows the lowest CT, 1,770K, with an EQE of 18.8 (28.0 lm/W, 39.8 cd/A) at 1,000 cd/m<sup>2</sup>. The reason why Device II-B exhibits the highest power efficiency, compare to other double emissive layer devices (Device II-A and Device II-C), with relatively low CT may be because the carriers have been most uniformly distributed into both EMLs. In Device II-A, most of the carriers concentrate in the orange red EML, resulting in a dominant orange-red emission. Blending some Spiro-2CBP into the BSB interlayer, Device II-B and II-C possesses a relatively low HOMO but a high LUMO, would create therein some channels for holes to freely penetrate through and block the entering electrons on the orange red EML to transport further into blue EML due to the electron barrier formed between Spiro-2CBP and Ir(2-phq)<sub>3</sub>. While the blend interlayer containing Device II-B and II-C reveals the current density lower than that of Device II-A, their balanced carriers (electrons and holes) modulation lead to high device efficiency and color stability. (see Fig. 2b)

## 4. Conclusion

In conclusion, a very low CT-OLED lighting has the great potential to diminish the adverse effect of lights on human physiology and psychology in the night time. This work demonstrated some highly efficient and very low CT-OLEDs devices using both single and double EML architecture. Single EML device has demonstrated the highest efficacy 31.7 lm/W (EQE 19.2% and current efficiency 42.9) with an incredible low CT 1,743K at 1,000 cd/m<sup>2</sup>. Because EML architecture favours the hole injection and enables excitons to form on the host. ECL modulate the excessive hole injection and confine the electrons within EML. Another hand the best double EML device has revealed the efficacy 29.1 lm/W (EQE 20.8%) with a CT 1,940K. The high efficiency may be attributed to use of EL efficient materials, efficient host to guest energy transfer and blue emission free light may help to achieve such a low CT. Thus the proposed both single and double EML based Device I-B and II-B may be an ideal lighting source for light at night to safeguard human health.

## 5. Acknowledgement

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## 6. References

- [1] C. W. Tang and S. A. VanSlyke, Organic electroluminescent diodes, *Appl. Phys. Lett.* 1987, **51** (12): 913-915.
- [2] J. Kido, M. Kimura and K. Nagai, Multilayer white light-emitting organic electroluminescent device, *Science*, 1995, **267** (5202): 1332-1334.
- [3] B. W. D'Andrade and S. R. Forrest, *Adv. Mater.* White organic light emitting diodes for solid-state lighting, 2004, **16** (18): 1585-1595.
- [4] S. F. Service, Organic LEDs looks forward to a bright, white future, *Science*, 2000, **310** (5755): 1762-1763.
- [5] F. So, J. Kido and P. Burrows, Organic light-emitting devices for solid state lighting, *MRS Bulletin* 2008, **33**, 663-669.
- [6] Y. Sun, N. C. Giebink, H. Kanno, B. Ma, M. E. Thompson and S. R. Forrest, Management of singlet and triplet excitons for efficient for white organic light emitting devices, *Nature* 2006, **440**, 908-912.
- [7] G. C. Brainard, B. A. Richardson, T. S. King and R. J. Reiter, The influence of different light spectra on the suppression of pineal melatonin content in the syrian hamster, *Brain Res.*, 1984, **294** (2): 333-339.
- [8] R. Kuller and L. Wetterberg, Melatonin, cortisol, EEG, ECG and subjective comfort in healthy humans: Impact of two fluorescent lamp types at two light intensities, *Lighting Res. and Tech.*, 1993, **25** (2): 71-81.
- [9] S.W. Lockley, G. C. Brainard and C. A. Czeisler, High sensitivity of the human circadian melatonin rhythm to resetting by short wavelength light, *J. Clin. Endocrinol. Metab.*, 2003, **88** (9): 4502-4505.
- [10] S. M. Pauley, Lighting for the human circadian clock: recent research indicates that lighting has become a public health issue, *Med. Hypotheses*, 2004, **63** (4) 588-596.

- [11] M. Sato, T. Sakaguchi and T. Morita, The effects of exposure in the morning to light of different color temperatures on the behaviour of core temperature and melatonin secretion in humans, *Biological Rhythm Research*, 2005, **36** (4): 287-292.
- [12] W. J. M. Van Bommel, Non-visual biological effect of lighting and the practical meaning for lighting for work, *Appl. Ergon.*, 2006, **37** (4): 461-466.
- [13] P. R. Mills, S. C. Tomkins and L. J. M. Schlangen, The effect of high correlated color temperature office lighting on employee wellbeing and work performance, *J. Circadian Rhythms*, 2007, **5**, 2.
- [14] M. T. Lee, C. H. Liao, C. H. Tsai and C. H. Chen, Highly efficient deep-blue doped organic light-emitting devices, *Adv. Mater.*, 2005, **17** (20): 2493-2497.
- [15] J. H. Jou, M. H. Wu, S. M. Shen, H. C. Wang, S. Z. Chen, S. H. Chen, C. R. Lin and Y. L. Hsieh, Sunlight-style color-temperature tunable organic light-emitting diode, *App. Phys. Lett.*, 2009, **95**, 013307.
- [16] S. Reineke, F. Lindner, G. Schwartz, N. Seidler, K. Walzer, B. Lussem, and K. Leo. White organic light emitting diodes with fluorescent tube efficiency, *Nature*, 2009, **459**, 234-238.
- [17] S. H. Eom, Y. Zheng, E. Wrzesniewski, J. Lee, N. Chopra, F. So and J. Xue, White phosphorescent organic light-emitting devices with dual triple-doped emissive layers, *Appl. Phys. Lett.*, 2009, **94**, 153303.
- [18] S. J. Su, E. Gonmori, H. Sasabe and J. Kido, Highly efficient organic blue-and white-light-emitting devices having a carrier- And exciton-confining structure for reduced efficiency roll-off, *Adv. Mater.*, 2008, **20** (21): 4189-4194.
- [19] V. I. Adamovich, S. R. Cordero, P. I. Djurovich, A. Tamayo, M. E. Thompson, B. W. D'Andrade and S. R. Forrest, New charge-carrier blocking materials for high efficiency OLEDs, *Org. Electron.* 2003, **4** (2): 77-87.
- [20] J. H. Jou, Y. S. Chiu, C. P. Wang, R. Y. Wang and H. C. Hu, Efficient, color-stable fluorescent white organic light-emitting diodes with single emission layer by vapor deposition from solvent premixed deposition source, *Appl. Phys. Lett.* 2006, **88**, 193501.
- [21] J. H. Jou, S. H. Chen, Shih M. Shen, Y. C. Jou, C. H. Lin, S. H. Peng, S. P. Hsia, C. W. Wang, and C. C. Chen, High efficiency low color-temperature organic light-emitting diode with a blend interlayer, *J. Mater. Chem.*, 2011, **21**, 17850-17854.
- [22] J. H. Jou, C. C. Chen, Y. C. Chung, M. F. Hsu, C. H. Wu, S. M. Shen, M. H. Wu, W. B. Wang, Y. C. Tsai C. P. Wang, and J. J. Shuye, Nanodot-enhanced high-efficiency pur white organic light emitting diode with mixed-host structures, *Adv. Funct. Mater.* 2008, **9**(1) 121-126.