Journal of
Photonics for Energy

SPIEDigitalLibrary.org/jpe

Candlelight style organic light-emitting diode: a plausibly human-friendly safe night light

Jwo-Huei Jou
Chun-Yu Hsieh
Po-Wei Chen
Sudhir Kumar
James H. Hong

SPIE

Candlelight style organic light-emitting diode: a plausibly human-friendly safe night light

Jwo-Huei Jou,* Chun-Yu Hsieh, Po-Wei Chen, Sudhir Kumar, and James H. Hong
National Tsing Hua University, Department of Materials Science and Engineering, Hsin-Chu 30013, Taiwan, China

Abstract. Candles emit sensationally warm light with a very low color temperature, comparatively most suitable for use at night. In response to the need for such a human-friendly night light, we demonstrate the employment of a high number of candlelight complementary organic emitters to generate and mimic candlelight based on organic light emitting diode (OLED). One resultant candlelight style OLED shows a very-high color rendering index (CRI), with an efficacy at least 300 times that of a candle or at least two times that of an incandescent bulb. The device can be fabricated, for example, by using four candlelight complementary emitters: red, yellow, green, and sky-blue phosphorescent dyes. These dyes, in the present system, can be vacuum deposited into two emission layers that are separated by a nanolayer of carrier modulation material that is used to maximize very high CRI and energy efficiency. A nano carrier modulation layer also played a significant role in maintaining the low blue emission and high-red emission, the low color temperature of device was obtained. Importantly, a romantic sensation giving and supposedly physiologically friendly candlelight style emission can hence be driven by electricity in lieu of hydrocarbon burning and greenhouse gas-releasing candles that were invented 5000 years ago. [DOI: 10.1117/1.JPE.4.043598]

Keywords: organic light-emitting diode; candlelight; chromaticity tunable; high-color rendering index and low-color temperature.

Paper 13039P received Nov. 28, 2013; revised manuscript received Jan. 14, 2014; accepted for publication Feb. 11, 2014; published online Apr. 11, 2014.

1 Introduction

Latest studies on light sources mainly focus on the development of energy-saving and environmentally friendly illumination devices.[1] Amongst, light-emitting diode (LED) and organic light-emitting diode (OLED) solid-state lighting[2–6] technologies have already achieved power efficiency (PE) near to that of fluorescent tubes. However, researchers have rarely focused on developing physiologically friendly light sources, especially for use at night. Color temperature (CT) of light plays an important role in human physiology and psychology.[7–14] Numerous studies have shown that high CT-lighting source and intensive white light or light with strong-blue emission drastically suppresses the secretion of melatonin (MLT), an oncostatic hormone. Importantly, the lack of MLT due to frequent exposure to intense light at night can increase the risk of breast, colorectal, and prostate cancers.[10] Suppression of MLT secretion has been reported upon exposure of 3000- or 5000-K fluorescent lights at 200 lx,[15] which is dimmer than the typical 500-lx office lighting, but brighter than the 100-lx lighting used at home.[10,15] Much milder suppression can only be observed as the CT is further reduced.

The importance of light sources with a low CT can be further realized by the fact that candlelight, which exhibits a CT around 1900 K is capable of creating romantic atmosphere during dinner time.[16] The pleasant sensation may originate from the naturally occurring MLT secretion, which helps people relax. In cases where lighting is needed, this secretion is less suppressed

*Address all correspondence to: Jwo-Huei Jou, E-mail: jjou@mx.nthu.edu.tw.

when dim light with a low CT is applied.[14,17] Although the low CT candles may be used as a physiologically friendly lighting measure at night, they are very energy-inefficient, not to mention their potential fire hazard problems, flickering nature, and unpleasant smoke due to incomplete burning. All the other hydrocarbon burning-based lighting devices, e.g., oil, kerosene, or gas lamps, are not energy-saving either.

Over the past 150 years, many electricity driven lighting technologies have emerged,[2–5] and energy efficient lighting tools have become possible. Lighting up the dark energy efficiently is no longer a major problem. However, these electrically driven lighting devices show no CT smaller than 2000 K. For example, the lowest CT is around 2500 K for incandescent bulbs, while 3000 or 5000 K for cold- or warm-white fluorescent tubes or compact fluorescent tubes, or 3000 or 5000 K for warm- or cold-white LED luminaires. They may be suitable for illumination during the daytime or at work, but apparently not for use at night. The true problems have actually arisen from the overuse of bright white light at night. Developing a new lighting source with low CT for reduced MLT suppression is hence no less urgent or less important than achieving an even higher lighting efficacy.

We demonstrate a candlelight style OLED device with a yellowish orange emission with Commission International de l'Eclairage (CIE) 1931 coordinates tunable around (0.52, 0.43) with a CT of 2000 K, closely matching the (0.52, 0.42) and 1914 K of a white candle studied. The resulting emissive spectrum shows an 80% similarity with that of the candle. The candlelight style OLED exhibits a 19-lm/W efficacy and a 93 color rendering index (CRI) while the efficacy is 0.1 lm/W and CRI 83 for candles. Furthermore, candlelight has a CT varying with the variation in emissive flame position, as seen in Fig. 1. The color temperature ranges from 1847 to 2626 K, with a 1914 K at the brightest spot.

2 Experiment

2.1 *Device Structures and Fabrication of Candlelight Style OLEDs*

Each device consisted of a 125-nm indium tin oxide layer, a 35-nm poly(3,4-ethylene-dioxy-thiophene)-poly(styrenesulfonate) (PEDOT:PSS) hole-injection layer, a 20-nm di-[4-(N, Nditolylamino)-phenyl]cyclohexane (TAPC) electron-confining layer, an 5-nm-short wavelength emissive-layer, a 15-nm-long wavelength emissive-layer, a 32-nm 1,3,5-tris(N-phenylbenzimidazol- 2-yl)benzene (TPBi) electron transporting layer, a 0.8-nm lithium fluoride (LiF) layer and a 150-nm aluminum layer; short wavelength emissive-layer is 4,4′,4′′-tri(N-carbazolyl)triphenylamine (TCTA) doped with 20% bis[3,5-difl uoro-2-(2-pyridyl)phenyl]-(2-carboxypyridyl) iridium(III) (Firpic). The long wavelength emissive-layer consisted of a 1,3,5-tris(N-phenylbenzimidazol-2-yl)benzene (TPBi) host doped with 12.5% tris(2-phenyl-pyridine) iridium (Ir(ppy)3)

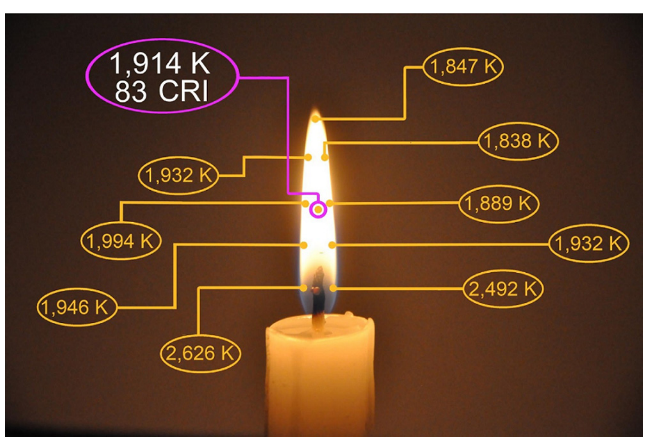

Fig. 1 Candle shows different color temperatures (CTs) at different positions inside the flame. The CT varies from 1847 to 2626 K for the white candle studied herein. To represent, the CT of the brightest spot is chosen, which is 1914 K (Ref. 18).

green dyes, 3% Iridium(III) bis(4-phenylthieno[3,2-c]pyridinato-N,C 2′)acetylacetonate (PO-01) yellow dye, and 1% bis(1-phenylisoquinolinolato-C2,N) iridium (acetylacetonate) ($Ir(piq)_2(acac)$) deep-red dye. The fabrication of the blend interlayer, short wavelength emissive layer and long wavelength emissive layer involved vapor deposition, and the sources were prepared via the solution premixing method.[19]

2.2 *Device Characterization*

The current-voltage-luminance characteristics of the resulting phosphorescent yellow OLEDs were measured using a Keithley 2400 electrometer together with a Minolta CS-100 luminance meter. Electroluminance (EL) spectrum and CIE color coordinates were obtained by using a PR 655 SpectraScan spectroradiometer.

3 Results and Discussion

Figure 2 shows the photographs of a red wine containing glass illuminated by different light sources that include the candlelight style OLED, a candle, a compact fluorescent tube, and a LED bulb. A yellowish orange emission was observed as the glass was shone by the candlelight style OLED, closely resembling that shone by the white candle. Whilst, the high CT compact fluorescent tube (5363 K) or LED bulb (5953 K) yielded a cold sensation by the white emission.

Table 1 summarizes the electroluminescent characteristics of the proposed devices. The EQE and PE of the device without an interlayer (Device I) is 16.3% and 30.3 lm/W with a CT of 2361 K and CRI of 64 at 1000 cd/m^2. With the adding of a 2-nm TCTA interlayer, the efficiency increased to 17.8% in EQE and 30.8 lm/W, and without a valuable CT and CRI. However, the blend interlayer with a 1:3 weight ratio of TCTA to TPBi in Device III markedly improved device CRI. Comparing with Device I, the CRI increased from 64 to 84, with a 12.5 lm/W in PE, 8.7% in EQE, and 3371 K in CT increase.

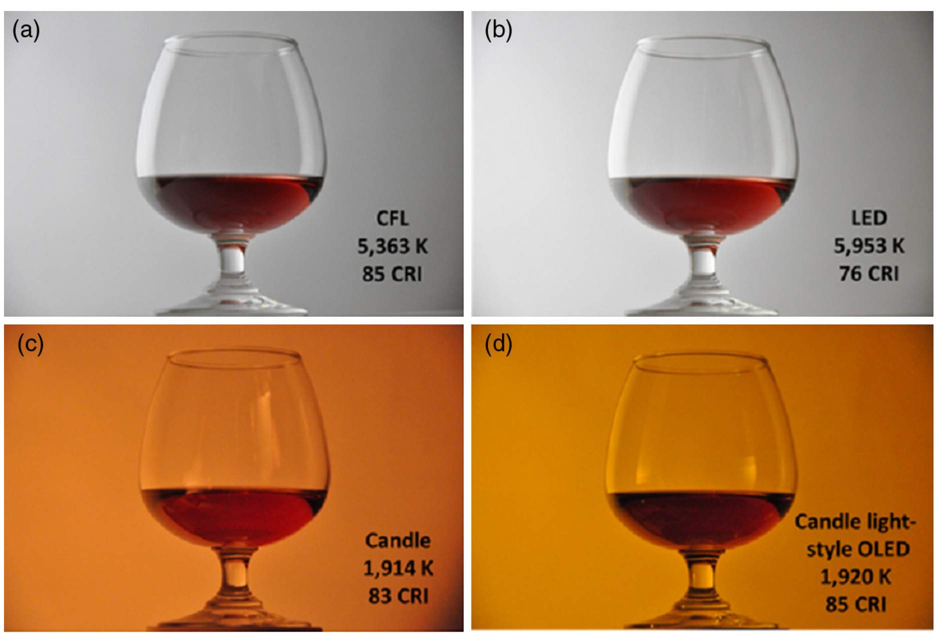

Fig. 2 Lighting color-temperature effect on the photographic results of a grape-wine containing glass illuminated under different light sources, including (a) a cold white compact fluorescent lamp (CFL) with 5363 K and an 85 color rendering index (CRI), (b) a white light-emitting diode (LED) bulb with 5953 K and a 76 CRI, (c) a candle with 1914 K and an 83 CRI, and (d) the candlelight-style OLED with 1920 K and an 85 CRI. Warm sensation is originated from the candle and the candlelight-style OLED that have relatively low CT, whereas cold sensation is generated from the high CT, CFL, and LED with white emission.[18]

Table 1 Effect of thickness and composition of a carrier modulation layer (CML) on the device color temperature (CT), candlelight spectrum resemblance, color rendering index (CRI), and power efficiency results (Ref. 18).

	CML			CT[a] [K]	CSR[b] [%]	CRI[a]	PE[a] [lm/W]	EQE[a] (%)
		Composition						
Device	THK[c] [nm]	TCTA	TPBi	at 100/1,000/10,000 cd/m²				
I	—	—	—	2,358/2,361/2,446	65.2	63/64/65	40.0/30.3/16.5	17.5/16.3/13.3
II	2	1	—	-/-/-	66.2	-/-/-	42.9/30.8/15.6	19.7/17.8/13.7
III	2	1	3	2,939/3,371/-	71.4	84/84/-	22.1/12.5/4.9	11.8/8.7/5.1
IV	2	3	1	2,050/2,150/2,450	80.3	86/93/93	29.6/19.2/9.4	17.4/14.6/10.6
V	2.5	3	1	2,065/2,257/2,622	79.9	88/93/90	26.3/15.6/7.0	15.6/12.3/8.1
VI	1.5	3	1	2,311/2,401/2,638	75.6	80/83/83	27.9/19.4/10.1	14.6/12.9/9.9

[a]at 100/1,000/10,000 cd/m²;
[b]compared with normalized spectrum;
[c]Abbreviations; THK-Thickncss;

The efficiency increased to 14.6% and 19.2 lm/W when the amount of TPBi further decreased in Device IV, and decreased to 12.3% and 15.6 lm/W as the interlayer increased to 2.5 nm for Device V. As the interlayer thickness decreased to 1.5 nm in device VI, the EQE decreased to 12.9%, although the PE of Device VI, which is 19.4 lm/W, is very similar to that of Device III. These three devices show comparatively high CRI, i.e., 93 for Device IV, 93 for Device V, and 83 for VI.

Device II exhibits the highest efficiency, and it may be attributed to the electrons confinement effect of the carrier modulation layer (CML), TCTA, due to its relative high LUMO level compared with that of TPBi and forms a significant barrier for electrons to overcome. The energy diagram in Fig. 3 can be used to illustrate this effect. As the use of TCTA as CML, a large amount

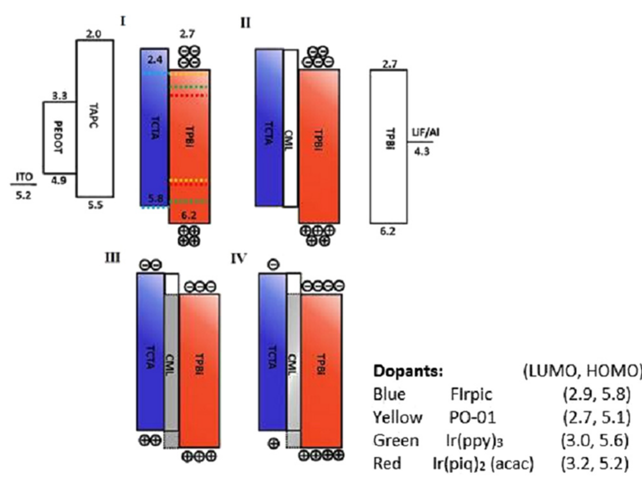

Fig. 3 Schematic illustration of candlelight style OLEDs without an interlayer (Device I), and with a neat TCTA interlayer (Device II). Devices III to VI all had a blend carrier modulation layer (CML), where the thickness was 1.5 nm for Device VI, 2.5 nm for Device V, and 2-nm Devices III and IV. The ratios of TCTA to TPBi in the blend layers of Devices II to VI were 1:0, 1:3, 3:1, 3:1, 3:1, respectively. This figure also shows the plausible distributions of holes and electrons in the studied devices at the same current density. With the use of a blend interlayer with proper compositions and thicknesses, the entering holes and electrons can be well distributed into the two-emissive zones.

of electrons are confined in the red and yellow emissive layer, which enables more excitons to generate and hence the device efficiency increased.

However, either with or without a CML, no blue emission can be observed from the EL spectrum, as Fig. 4. To enable more electrons to inject into the blue-emissive layer, the TCTA-based CML was modified by blending it with an electron-transporting material TPBi, via which electrons can much more freely penetrate from the yellow red emissive layer to blue emissive layer. This enhances the intensity of the blue emission. Although, the blue emission was overly increased due to the presence of a new barrier (0.4 eV) against holes at the interface between the blue emissive layer and the TPBi in the blend CML, and causes too few holes to enter the yellow red emissive zone.

To enable sufficient holes to inject into the yellow red emissive layer, blend CML was further modified by reducing the amount of TPBi used, which is also effective in electron transporting as mentioned before. This explains why the blue emission markedly decreased when the ratio of TCTA: TPBi employed fell from 1:3 to 3:1, as shown in Fig. 5.

The function of the blend CML is also sensitive to the employed thickness. Increase or decrease the thickness of CML, both can lower the device efficiency. This may be because more excitons were generated within the CML, either on TCTA or on TPBi. Although energy transfer can still occur via diffusion of the generated excitons, their effectiveness drastically decreased as the diffusion distance increased, which explains why both the intensities dropped as the CML thickness increase. In case of thinner CML thickness device, no sufficient excitons are generated and diffuse into emissive zone, cause a lower efficiency.

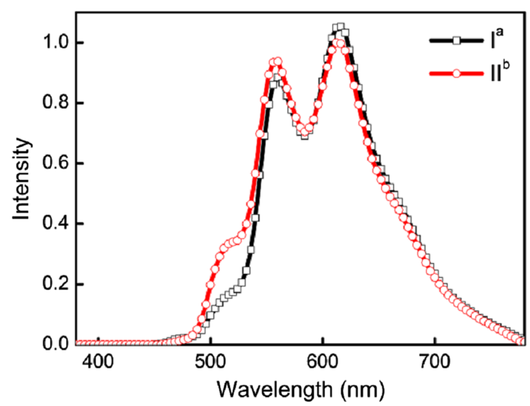

Fig. 4 Electroluminescent spectra of no and neat CML-composing counterparts. (a) With no CML, (b) a 2-nm neat TCTA as interlayer. All spectra were obtained at 5 A/m^2.

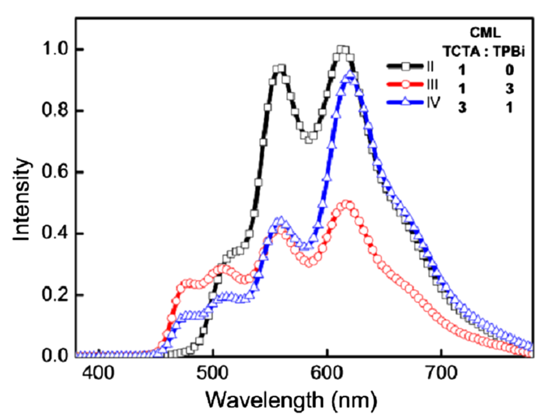

Fig. 5 Electroluminescent spectra of the blend CML-composing devices.

4 Conclusion

In this study, we demonstrate a candlelight style OLED device with a yellowish orange emission with Commission International de l'Eclairage (CIE) 1931 coordinates tunable around (0.52, 0.43) with a CT of 2000 K, closely matching the (0.52, 0.42) and 1914 K of a white candle studied. The candlelight style OLED exhibits a 19 lm/W efficacy and a 93 CRI, while the efficacy is 0.1 lm/W and CRI 83 for candles. The high efficiency of the proposed device may be attributed to the employed blend interlayer, which helps effectively distribute the entering carriers into the available recombination zones. These candlelight style OLEDs are an ideal lighting source to safeguard human health, especially for use at night.

Acknowledgments

The authors would like to acknowledge support from National Science Council and Ministry of Economic Affairs, Taiwan. This work was financially supported in part by grants: NSC101-3113-E-007-001, NSC100-2119-M-007-011-MY3, and MEA 100-EC-17-A- 07-S1-181.

References

1. V. Adamovich et al., "High efficiency single dopant white electrophosphorescent light emitting diodes," *New J. Chem.* **26**(9), 1171–1178 (2002), http://dx.doi.org/10.1039/b204301g.
2. J. Kido, M. Kimura, and K. Nagai, "Multilayer white light-emitting organic electroluminescent device," *Science* **267**(5202), 1332–1334 (1995), http://dx.doi.org/10.1126/science.267.5202.1332.
3. R. F. Service, "Organic LEDs look forward to a bright, white future," *Science* **310**(5755), 1762–1763 (2005), http://dx.doi.org/10.1126/science.310.5755.1762.
4. F. So, J. Kido, and P. Burrows, "Organic light-emitting devices for solid-state lighting," *MRS. Bull.* **33**(7), 663–669 (2008), http://dx.doi.org/10.1557/mrs2008.137.
5. J. H. Jou et al., "High-efficiency flexible white organic light-emitting diodes," *J. Mater. Chem.* **20**(32), 6626–6629 (2010), http://dx.doi.org/10.1039/c0jm01348j.
6. J. H. Jou et al., "Sunlight-style color-temperature tunable organic light-emitting diode," *Appl. Phys. Lett.* **95**(1), 013307 (2009), http://dx.doi.org/10.1063/1.3176217.
7. W. J. M. van Bommel, "Non-visual biological effect of lighting and the practical meaning for lighting for work," *Appl. Ergon.* **37**(4), 461–466 (2006), http://dx.doi.org/10.1016/j.apergo.2006.04.009.
8. L. W. R. Küller, "Melatonin, cortisol, EEG, ECG and subjective comfort in healthy humans: Impact of two fluorescent lamp types at two light intensities," *Light. Res. Technol.* **25**(2), 71–80 (1993), http://dx.doi.org/10.1177/096032719302500203.
9. P. R. Mills, S. C. Tomkins, and L. J. M. Schlangen, "The effect of high correlated colour temperature office lighting on employee wellbeing and work performance," *J. Circadian Rhythms* **5**(2) (2007), http://dx.doi.org/10.1186/1740-3391-5-2.
10. S. M. Pauley, "Lighting for the human circadian clock: recent research indicates that lighting has become a public health issue," *Med. Hypotheses* **63**(4), 588–596 (2004), http://dx.doi.org/10.1016/j.mehy.2004.03.020.
11. G. C. Brainard et al., "The Influence of various irradiances of artificial-light, twilight, and moonlight on the suppression of pineal melatonin content in the syrian-hamster," *J. Pineal. Res.* **1**(2), 105–119 (1984), http://dx.doi.org/10.1111/jpi.1984.1.issue-2.
12. 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.* **88**(9), 4502–4505 (2003), http://dx.doi.org/10.1210/jc.2003-030570.
13. F. A. J. L. Scheer and R. M. Buijs, "Light affects morning salivary cortisol in humans," *J. Clin. Endocrinol. Metab.* **84**(9), 3395–3398 (1999), http://dx.doi.org/10.1210/jcem.84.9.6102.
14. M. Sato, T. Sakaguchi, and T. Morita, "The effects of exposure in the morning to light of different color temperatures on the behavior of core temperature and melatonin secretion in

humans," *Biol. Rhythm Res.* **36**(4), 287–292 (2005), http://dx.doi.org/10.1080/09291010500079734.
15. T. Kozaki et al., "Effects of short wavelength control in polychromatic light sources on nocturnal melatonin secretion," *Neurosci. Lett.* **439**(3), 256–259 (2008), http://dx.doi.org/10.1016/j.neulet.2008.05.035.
16. C. Potera, "The core of the candle problem," *Environ. Health Perspect.* **108**(4), A165–A165 (2000).
17. A. Panzer, "Depression or cancer: the choice between serotonin or melatonin," *Med. Hypotheses* **50**(5), 385–387 (1998), http://dx.doi.org/10.1016/S0306-9877(98)90209-2.
18. J. H. Jou et al., "Candle light-style organic light-emitting diodes," *Adv. Funct. Mater.* **23**(21), 2750–2757 (2013), http://dx.doi.org/10.1002/adfm.v23.21.
19. J. H. Jou et al., "Efficient, color-stable fluorescent white organic light-emitting diodes with single emission layer by vapor deposition from solvent premixed deposition source," *Appl. Phys. Lett.* **88**(19), 193501 (2006), http://dx.doi.org/10.1063/1.2200007.

Biographies of the authors are not available.