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Candle Light-Style Organic Light-Emitting Diodes

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In response to the call for a physiologically-friendly light at night that shows low color temperature, a candle light-style organic light emitting diode (OLED) is developed with a color temperature as low as 1900 K, a color rendering index (CRI) as high as 93, and an efficacy at least two times that of incandescent bulbs. In addition, the device has a 80% resemblance in luminance spectrum to that of a candle. Most importantly, the sensationally warm candle light-style emission is driven by electricity in lieu of the energy-wasting and greenhouse gas emitting hydrocarbon-burning candles invented 5000 years ago. This candle light-style OLED may serve as a safe measure for illumination at night. Moreover, it has a high color rendering index with a decent efficiency.

1. Introduction

Candles were invented 5000 years ago and are today still used extensively. Candle light is able to create a romantic atmosphere,[1] and the pleasant sensation may originate from the naturally occurring melatonin secretion,[2] which helps people relax. In cases where lighting is needed, this secretion is less suppressed when dim light with a low color temperature is applied.[3] In contrast, melatonin generation would be markedly suppressed in the presence of a bright light with a high color temperature.[4–8] Importantly, the lack of melatonin due to frequent exposure to intense light at night can increase cancer risk.[4–6] Suppression of melatonin secretion has been reported upon exposure of 3000 or 5000 K fluorescent lights at 200 lx,[9] which is dimmer than the typical 500 lx office lighting, but brighter than the 100 lx lighting used at home.[4,9] Much milder suppression can only be observed as the color temperature is further reduced. The notorious effect of blue light on melatonin suppression has also been identified.[10] These studies support calls from medical experts for the development of a new light source for use at night that shows low color temperature or is free of blue emission to safeguard human health.[4,9]

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DOI: 10.1002/adfm.201203209

Although the low color temperature 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,[11–14] and energy-efficient lighting tools have become possible. Lighting up the dark energy-efficiently is no longer a major problem. The true problems have actually arisen from the overuse of bright white light at night. Over the past decades, high-efficiency light emitting diodes (LEDs)[15–19] and organic light emitting diodes (OLEDs)[20–25] had been developed. White LEDs with 209 lm W^{-1} chip efficacy, or 67 lm W^{-1} luminaire efficacy have been reported.[26] White OLEDs with 33 to 124 lm W^{-1} efficacy have been reported for laboratory studies,[20] while 11 to 25 lm W^{-1} have been achieved for the first generation commercial OLED lighting panels. These solid-state lighting devices have demonstrated approaching fluorescent lamp efficacy. Nevertheless, all the electricity-driven luminaires are either cool- or warm-white with a color temperature much greater than 2500 K. They may be suitable for illumination during the daytime or at work, but apparently not for at night. Developing a new lighting source with low color temperature for reduced melatonin suppression is hence no less urgent or less important than achieving an even higher lighting efficacy. However, little attention had been paid to this until very recently. In 2009, a sunlight-style OLED was reported with a color temperature tunable from 2300 to 12 000 K,[27] proving the possibility in fabricating a new light source with a low color temperature using OLED rather than LED technology. This is because the intense blue emission driven LED lighting panels would become energy-inefficient if the blue light were removed or significantly diminished in order to obtain a low color temperature. The comparison studies performed here were carried primarily for comparing the low color temperature OLEDs with the conventional blue III-nitride LEDs integrated with phosphor technology. It is important to note that the III-Nitride LEDs[28–34] are very high efficacy emitters for solid state lighting in comparison to OLEDs. Recent works on III-Nitride LEDs have also focused on the pursuit of high-efficiency active regions based on semi-/nonpolar quantum wells[28,29] and polar quantum wells with large overlap designs[30–34] for achieving high-performance nitride-based LEDs emitting in the longer visible spectral range specifically in the green up to red spectral regimes. Meanwhile, OLEDs with numerous monochromatic as well as polychromatic emissions can be made energy-efficiently and achieve a high color rendering index (CRI). Low color temperature OLEDs with high device efficiency and CRI can hence be made.

In 2011, low color temperature OLEDs with color temperature ranging from 1800 to 2800 K were reported with 30 lm W^{-1} at 1000 cd m^{-2} without using any light extraction method.[35] Typically, device efficacy can be magnified to two or three times if effective internal and external light extraction structures are coupled. Although the bare device efficacy was sound, the color rendering quality was low. In 2012, a low color temperature OLED with a high CRI of 87 was presented.[36] However, the device showed an 8.6 lm W^{-1} efficacy, much lower than that of incandescent bulbs. The current challenge has therefore become how to devise a lighting source having simultaneously a very-low color temperature, a high power efficiency, and a very high CRI to ensure the resulting illumination be aesthetically pleasing and physiologically-friendly, besides being energy-saving.

We present in this report a candle light-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 color temperature of 2000 K, closely matching the (0.52, 0.42) and 1914 K of a white candle studied. The resulting emissive spectrum shows an 81% similarity with that of the candle (**Figure 1**). The candle light-style OLED exhibits a 19 lm W^{-1} efficacy and a 93 CRI, while the efficacy is 0.1 lm W^{-1} and CRI 90 for candles. Figure 1a compares the experimentally measured power spectrum of the candle light-style OLED with that of the white candle. These two light sources could hardly be thought similar by looking at their power distribution spectra. Nevertheless, they both showed the same trend, with the emissive intensity increasing with the increase of wavelength. Additionally, infrared shows no contribution in viewed effect to human eyes, and deep-red is weak in luminosity. As a result, the extremely large disparity emission in the long wavelength visible range would have little effect visually. In contrast, by converting the power into luminance distribution, that was done by convoluting the entire power spectrum with the luminosity function, the two light sources showed a 80% similarity, as seen in Figure 1b. The resultant luminance spectrum of the candle light-style OLED peaked at 560 and 608 nm, which respectively corresponded to a yellowish green light and an orange-red light. The combination of both led to a yellowish orange emission, matching that of the candle light peaking at 580 nm.

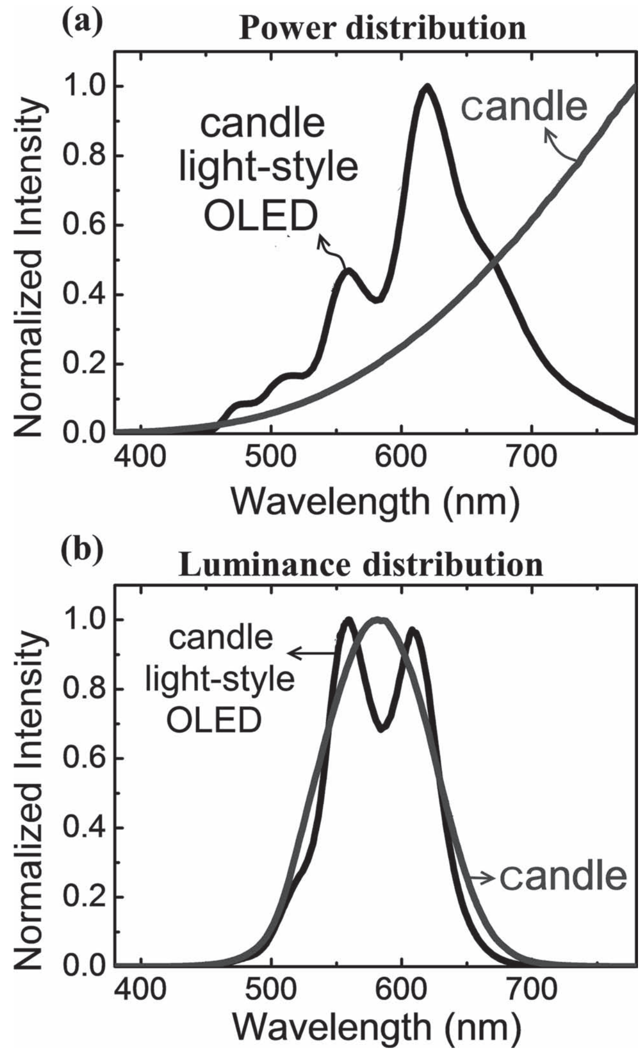


Figure 1. Comparison of the a) spectral power and b) luminosity distributions of the candle light-style OLED with the candle. Although the power spectra of the two light sources are quite dissimilar, they look almost the same from the viewpoint of human eyes. The candle light-style OLED shows an 80% similarity with the candle as comparing their spectral luminance distributions that are obtained by convoluting the power spectra with the luminosity function.

2. Results and Discussion

As noted, candle light is soft and smooth, having a continuous full spectrum. The emission is the strongest in the long wavelength region, and decreases monotonically toward the blue emission region. To generate a candle light-style emission, the OLED devices are designed to maximize the emission of the employed red light-emitting dye and minimize that of the blue counterpart accordingly. Since candles emit a tremendously high amount of infrared that cannot be visualized by human eyes and show no contribution in chromaticity, the candle light-style chromaticity is obtained by creating a spectrum mimicking that of candles on basis of the human eye's perspective. Furthermore, candle light has a color temperature varying with the variation of flame position, as seen in **Figure 2**. The color temperature ranges from 1847 to 2626 K, with 1914 K at the brightest spot.

The candle light-style OLEDs are made of four organic electro-phosphorescence dyes with red, yellow, green, and sky-blue colors, as shown in **Figure 3**. These four light-emitting dyes are dispersed in properly engineered hosting materials, and then deposited into two different emissive layers with a total thickness of 20 nm. To attain high device efficiency, the

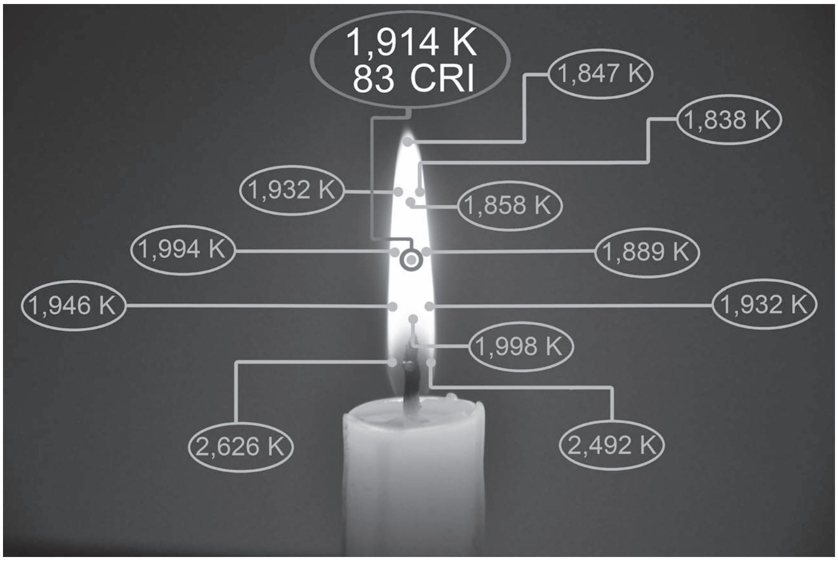


Figure 2. Candle shows different color temperatures at different flame positions. The color temperature varies from 1847 to 2626 K for the white candle studied. To represent this most effectively, the color temperature of the brightest spot is chosen, which is 1914 K.

emissive layers are sandwiched by two thin layers of light-emitting-auxiliary materials to facilitate the transport of carriers, and which are further sandwiched by two additional carrier injection layers to minimize the interfacial barrier in between the organic molecules and electrodes. A thin layer of aluminum is deposited as the cathode, and a transparent conductive oxide, indium tin oxide (ITO), is used as the anode.

Table 1 compares the color temperature, CRI, and efficacy of the candle light-style OLED with those of the cold-white fluorescent tube, cold-white LED bulb, cold-white compact fluorescent lamp, incandescent bulb, and white candle studied. While those cold-white lighting devices show color temperature as high as 6000 K, their warm-white counterparts can also show comparatively low color temperatures, mostly in the vicinity of 3000 K. The approach presented in this study may be extended to LEDs, and hence candle light-style LED may also be devised, taking its inherently energy-saving and high reliability advantages. However, nearly no lighting measures except for the OLED are energy-saving, and meanwhile being environmentally-friendly (e.g., mercury-free), human-friendly (e.g., flickering-, glare-, and UV-free), and and smoke- and fire hazard-free, as compared with candles. The candle light-style OLEDs presented may serve as an ideal physiologically-friendly lighting device for use at night, or an alternative romantic table light for dinner time.

The candle light-style OLED has two additional features, besides being energy-efficient and high color rendering. First, its color temperature can easily be tuned lower, such as 1800 K, by further diminishing the melatonin suppression sensitive blue emission, making which a physiologically more friendly lighting source than candles. Second, these candle light-style OLEDs yield no emission beyond the infrared region, hence giving a physically 'cold' glow, but with warm sensation.

Figure 4 shows the photographs of a red wine containing glass illuminated under different light sources that include the candle light-style OLED, a candle, a compact fluorescent tube, and an LED bulb. A yellowish orange emission was observed as the glass was irradiated by the candle light-style OLED with a color temperature of 1920 K and 85 CRI, close to that when a white candle was used (1914 K and 83 CRI). In addition, the high color temperature compact fluorescent tube (5363 K) or LED bulb (5953 K) yielded a cold sensation due the white emission.

As a good alternative to a candle, a candle light-style OLED is reliable, dimmable, and "invisible". In addition, it is reliable; unlike a candle flame, the OLED light is unaffected by wind. Also, a candle light-style OLED is dimmable; when it dims, along with a plane-type diffusive light, it reduces the unpleasant glare. Thus, because it is dimmable, the magnitude of the glare

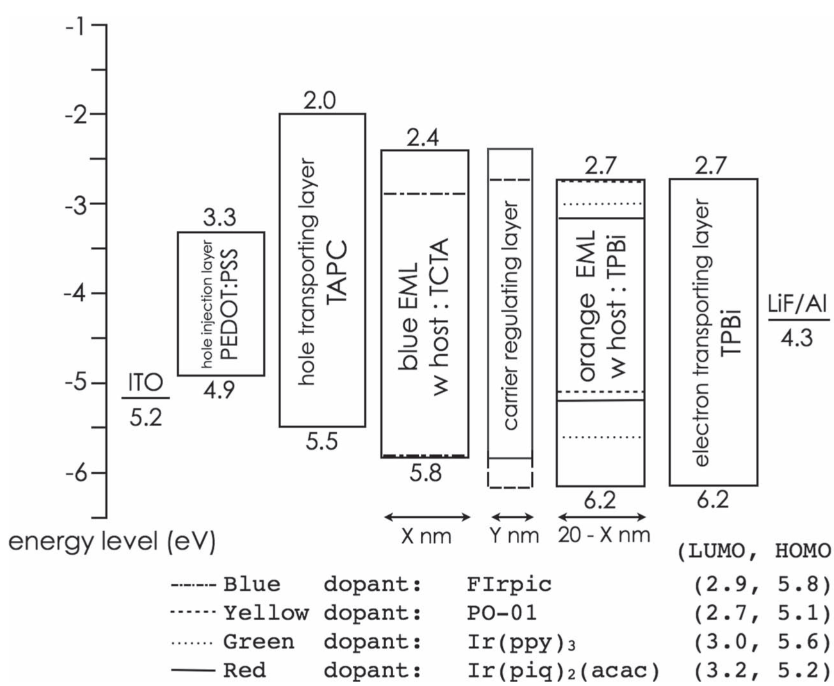
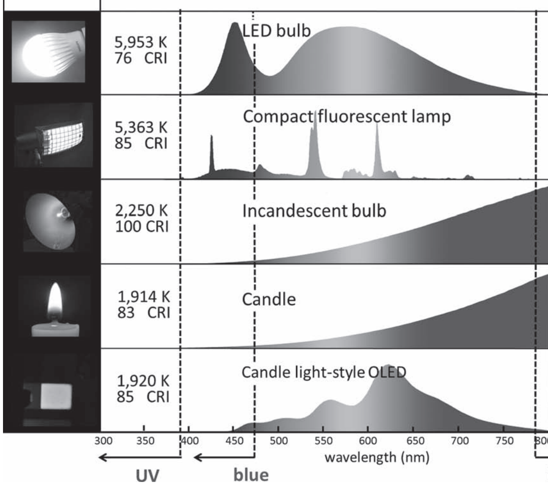


Figure 3. Schematic illustration of the candle light-style OLED device that composes four blackbody-radiation complementary dyes, namely red, yellow, green, and sky-blue, dispersed in two emissive layers separated by a nano-interlayer to harvest the ultimate color rendering index and device efficacy.

Table 1. Comparison of the candle light-style OLED with current lighting sources in terms of power spectrum, color temperature, color rendering index (CRI), and characteristics regarding mercury (Hg), flickering, glare, and ultraviolet (UV) and infrared (IR) emission. The presented candle light-style OLED is a promising candidate to echo the call of physicians for a blue emission free or low color temperature light source for the illumination at night to safeguard human health, since it contains little blue emission, and is low in color temperature, high in CRI, and free from Hg/flickering/glare/UV/IR.



Light source	Spectrum		Free of				
			Hg	Flicker-ing	Glare	UV	IR
	5,953 K 76 CRI	LED bulb	yes	yes	no	no	yes
	5,363 K 85 CRI	Compact fluorescent lamp	no	no	no	no	yes
	2,250 K 100 CRI	Incandescent bulb	yes	yes	no	yes	no
	1,914 K 83 CRI	Candle	yes	no	no	yes	no
	1,920 K 85 CRI	Candle light-style OLED	yes	yes	yes	yes	yes

produced by the OLED in **Figure 5**a can become significantly reduced. On the contrary, the glare from the candle flame in Figure 5 is unadjustable. In addition, a candle light style OLED can become "invisible" or away from a person's vision, when it is attached to a ceiling, while candles or candle lamps on a table remain visible. Although a candle lighting device can be attached to a ceiling, a distance between candle flames and the ceiling must be kept to avoid fire hazards. Although, from above, a candle light style OLED attached to a ceiling, shines its light upon all items in its range, the OLED can become selective in brightening objects because its light is directional according to Figure 5b.

Surrounded by darkness, the wine containing glass and the air above it are bright; this phenomenon in Figure 5b (right) shows that light from a candle light-style OLED can be single-sided emitting and hence be less obtrusive. This enables the OLED to offer more possible outcomes than an obtrusive candle, which releases its light to its environment in all directions. In contrast to an omni-directional candle light, both "directional" and omni-directional light can be emitted by the OLED, generating larger varieties to meet different lighting demands with changing fixture angles or positions.

Figure 6 compares the resulting color temperature, CRI, and power efficiency of the studied candle light-style OLED against those of the previously reported OLED devices and current

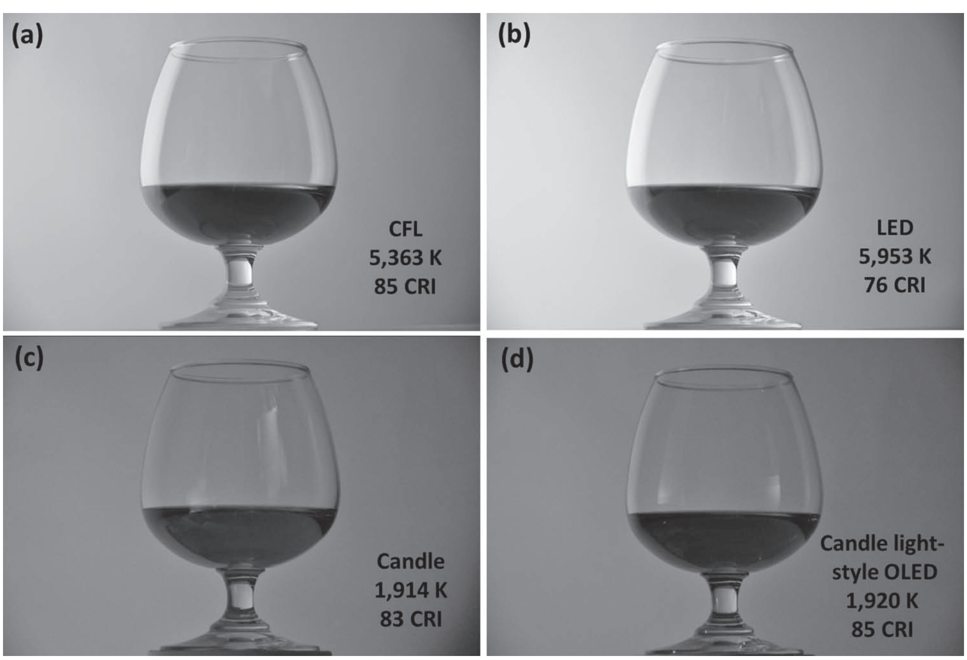


Figure 4. Lighting color temperature effect on the photographic results of a wine containing glass illuminated under different light sources, including a) a cold white compact fluorescent lamp (CFL) with 5363 K and an 85 CRI, b) a white LED bulb with 5953 K and a 76 CRI, c) a candle with 1914 K and an 83 CRI, and d) the candle light-style OLED with 1920 K and an 85 CRI. A warm sensation is perceived from the candle and the candle light-style OLED that have relatively low color temperature, while a cold sensation is perceived from the high color temperature CFL and LED with white emission.

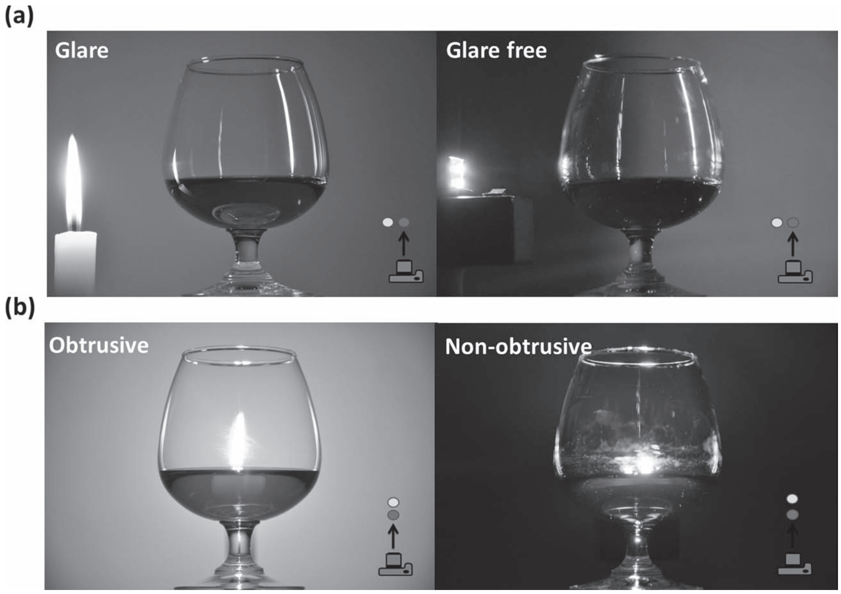


Figure 5. Dimmable and single-sided emitting characters of the candle light-style OLED warrant a lighting atmosphere free from a) glare and b) obtrusion. The photographs were taken by placing the light sources, the candle (left) and the candle light-style OLED (right), at positions a) beside, and b) right behind the wine containing glass.

luminaires, including an LED, a compact fluorescent tube, an incandescent bulb, and a candle. As seen, most of the efforts had been placed on developing efficient and/or very high CRI white lighting sources with a color temperature greater than 2500 K. Several low color temperature OLEDs had been reported, but lacking of either high CRI or high efficacy.[20,21,35–45] This is a low color temperature light source based on OLED that is demonstrated with a very high CRI and with a more than two times incandescent bulb efficacy. In addition, recent works have reported the use of 2D defective grating method for achieving improved light extraction and power efficiency in OLEDs[46] based on close-packed colloidal-based deposition[47,48] and colloidal-based imprinting method[49] The efficacy can be further doubled or tripled provided internal and external light extraction structures as well as high refractive glass are coupled.[20,50–52]

Table 2 summarizes the electroluminance characteristics of the devices studied. To attain the maximum CRI of the four-spectrum candle light-style OLED, the thickness of the emissive layers and doping concentration of the light-emitting dyes are optimized. However, the best CRI result is only 85 with a 17 lm W^{-1} in efficacy and 1900 K in color temperature without carrier regulating. By incorporating a 2-nm carrier regulating layer in between the two emissive layers, the CRI is increased to 93 and efficacy increased to 19 lm/W, but at the cost of increasing the color temperature from 1900 to 2100 K. The regulating layer is used to control the flow of holes, so that some holes can be retained in the blue emissive layer to yield the blue light needed for composing a high color rendering character. For this reason, the entering holes are purposely distributed into two emissive layers, instead of concentrating in one, leading to an increase in the device efficiency since more effective carrier recombination can take place in a wider emissive zone.

As also shown in **Figure 7**, the power efficiency of the OLEDs decreased significantly at high injection current level. The efficacy roll-off may be attributed to numerous factors, including concentration quenching,[53,54] exciton quenching,[54,55] imbalance carrier injection,[56] and ineffective carrier confinement.[57] To improve this, employing much diluted emitter,[58] using multiple emission layers with stepwise energy-levels to extend exciton generation zone or enabling excitons to generate on both host and guest,[59] improving the injection of minor carrier to balance the carriers,[60] and incorporating effective electron and/or hole confinement layers[21,61] have been found to be effective. These approaches may be adopted to further improve the efficacy of the present devices.

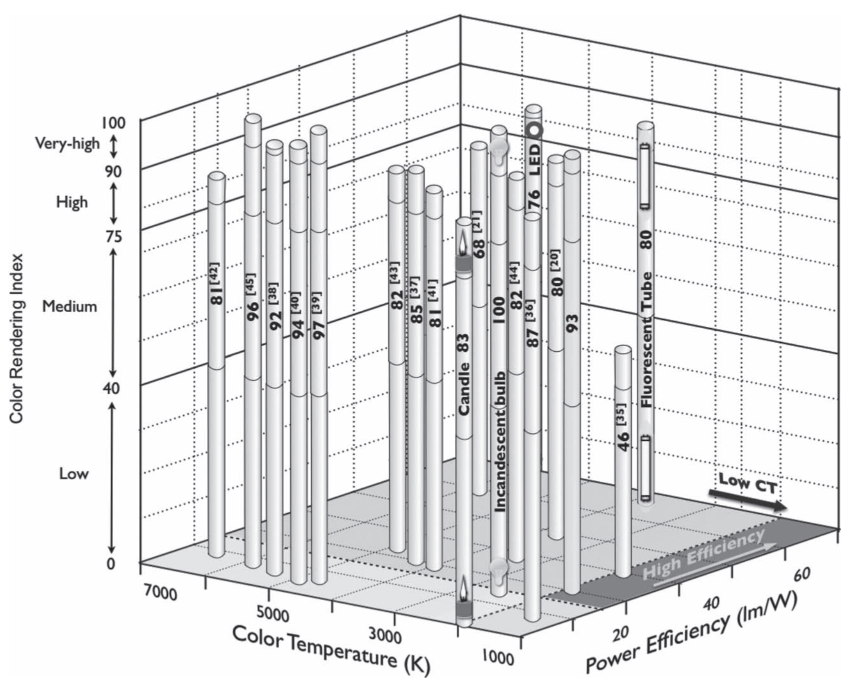


Figure 6. The candle light-style OLED shows a very high CRI (93), warranting a good visual quality, an at least greater than two times incandescent bulb efficacy, proving to be energy saving, and, most importantly, a very low color temperature (2150 K), enabling its derived lighting fixture a most physiologically-friendly lighting device ever for use at night.[20,21,35,45]

Table 2. Effect of the thickness (denoted THK) and dopant concentration of two emissive layers (EMLs) as well as thickness and composition of a carrier regulating layer (CRL) on the device color temperature (CT), candlelight spectrum resemblence (CSR), color rendering index (CRI), and power efficiency (PE) results.

Device	First EML		Second EML				CRL			CT [K][a)]	CSR [%][b)]	CRI[a)]	PE [lm w^{-1}][a)]
	THK [nm]	Dopant Concentration [wt%]	THK [nm]	Dopant Concentration [wt%]			THK [nm]	Composition					
		Blue		Green	Yellow	Red		TCTA	TPBi				
I-1	15	20	5	12.5	5	1	–	–	–	2900/3000/3200	69.8	78/78/80	18.9/15.9/7.8
I-2	10	20	10	12.5	5	1	–	–	–	1851/1918/2101	77.1	83/85/86	23.0/17.0/9.8
I-3	5	20	15	12.5	5	1	–	–	–	2358/2361/2446	65.2	63/64/65	40.0/30.3/16.5
II-1	5	20	15	12.5	5	1	2	3	1	2150/2300/2570	72.8	75/79/80	31.8/21.5/11.4
II-2	5	20	15	12.5	5	1.5	2	3	1	1970/2190/2510	79.0	83/90/89	24.2/16.1/7.7
II-3	5	20	15	12.5	4	1	2	3	1	2138/2187/2438	71.8	74/78/80	30.7/20.5/10.3
II-4	5	20	15	12.5	3	1	2	3	1	2050/2150/2450	80.3	86/93/93	29.6/19.2/9.4
II-5	5	20	15	10	3	1	2	3	1	1998/2118/2374	79.1	87/92/92	29.6/18.6/8.7
II-6	5	20	15	15	3	1	2	3	1	2508/2575/2853	74.6	77/80/80	37.1/24.4/12.1
II-7	5	20	15	12.5	3	1	2	1	1	2435/2710/2963	76.8	86/89/87	28.5/16.0/6.6
II-8	5	20	15	12.5	3	1	2	1	3	2939/3371/-	71.4	84/84/-	22.1/12.5/4.9
II-9	5	20	15	12.5	3	1	2	1	–	-/-/-	66.2	-/-/-	42.9/30.8/15.6
III-1	5	20	15	12.5	3	1	1.5	3	1	2311/2401/2638	75.6	80/83/83	27.9/19.4/10.1
III-2	5	20	15	12.5	3	1	2.5	3	1	2065/2257/2622	79.9	88/93/90	26.3/15.6/7.0

[a)]@100/1,000/10,000 cd m^{-2}; [b)]compared with normalized spectra.

3. Conclusion

We have demonstrated the first candle light-style lighting source based on a four-spectrum OLED. The resulting OLED exhibits a candle light-style yellowish orange chromaticity with an efficacy of 29 lm W^{-1} at 100 cd m^{-2} or 19 lm W^{-1} at 1000 cd m^{-2}, with a respective CRI of 87 or 93 and color temperature of 2050 or 2150 K. The infrared free low color temperature OLED delivers

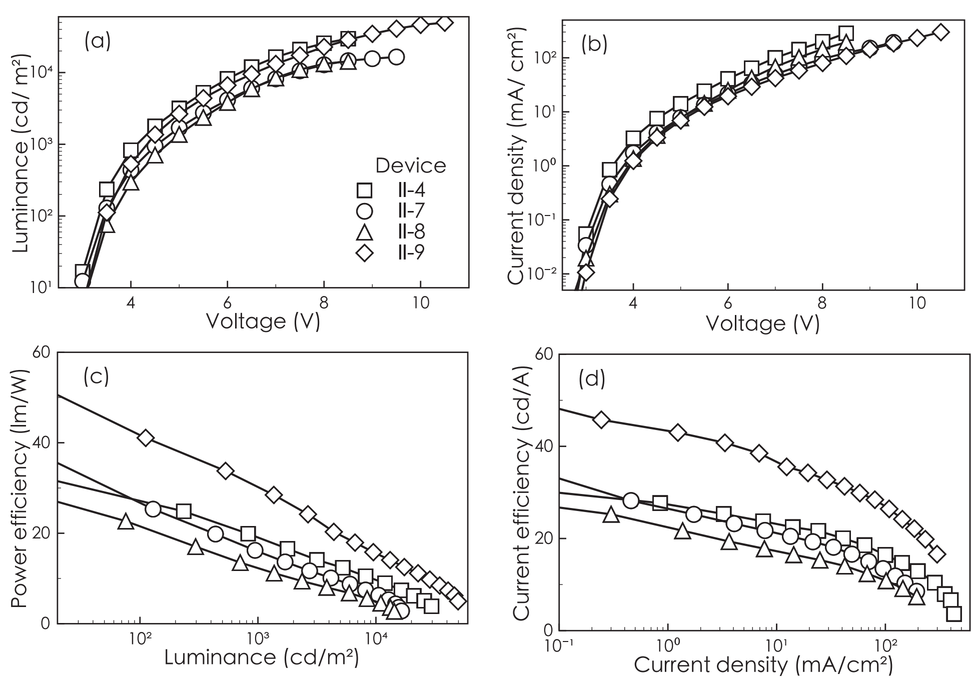


Figure 7. Effect of different composition of the blend interlayer on the resulting a) luminance, b) current density, c) power efficiency, and d) current efficiency of the candle light-style OLED.

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a sensationally warm but irradiation cold glow, which is energy-saving, poses no fire hazard, and is free from flickering, glare, and ultraviolet emission. In addition, the candle light-style OLED is more reliable, less obtrusive, or more "glare free" than a candle, providing its users more options in designing more human-friendly lighting fixtures; by doing so, this may lead them to a brighter future.

4. Experimantal Section

Device I-1 to I-3 consisted of a 125 nm indium tin oxide (ITO) layer, a 35 nm poly(3,4-ethylene-dioxy-thiophene)-poly(styrenesulfonate) (PEDOT:PSS) hole-injection layer (HIL), a 20 nm di-[4-(N,N-ditolylamino)-phenyl]cyclohexane (TAPC) electron-confining layer, an X (5,15,20) nm short wavelength emissive-layer, a 20-X nm long wavelength emissive-layer, a 32 nm 1,3,5-tris(N-phenylbenzimidazol-2-yl)benzene (TPBi) electron transporting layer (ETL), 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-difluoro-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 ($Ir(ppy)_3$) green dyes, 5% 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. Device II-1 to II-6 were based on Device I-3 and further incorporated with a 2 nm blend interlayer which composed of TCTA and TPBi between the two emissive-layers. In addition, the doping concentration effect of green, yellow, and red dyes in TPBi host were investigated separately. Different TCTA and TPBi composition ratio of blend interlayer refer to Device II-4 was studied in Device II-7, II-8, and II-9. The thickness effect on the interlayer was studied in Device III-1 and III-2. The fabrication of the blend interlayer, short wave length emissive-layer and long wave length emissive-layer involved vapor-deposition, and the sources were prepared via the solution premixing method.[62]

The fabrication process initially involved spin-coating an aqueous solution of PEDOT: PSS at 4000 rpm for 20 s to form the 35 nm HIL. The blend interlayer, EML and ETL were deposited using the thermal deposition method in a high vacuum chamber (3×10^{-5} Torr) at a rate of 0.7–1.5 Å s^{-1}. The electron injection layer LiF and aluminum cathode were also deposited using the thermal deposition method in a high vacuum chamber (3×10^{-5} Torr) at respective rates of 0.1 and 12 Å s^{-1}.

The material TPBi, TAPC, FIrpic, $Ir(ppy)_3$, $Ir(piq)_2(acac)$, and PO-01 were purchased from Luminescence Technology Corporation. The material TCTA was purchased from e-Ray Optoelectronics Technology. PEDOT:PSS (Clevios P VPAI4083) was purchased from Bayer Corporation. The LiF was purchased from Strem Chemicals Incorporation. The aluminum cathode was purchased from Showa Chemical Industry Company Ltd.

The luminance, spectrum and CIE chromatic coordinates results were measured by using a PR-655 spectroradiometer, and a Keithley 2400 electrometer was used to measure the current–voltage (*I–V*) characteristics.

Acknowledgements

This work was financially supported by Taiwan National Science Council and Ministry of Economic Affairs through grant numbers MEA 100-EC-17-A-07-S1-181, NSC 101-3113-E-007-001, NSC 100-2119-M-007-011-MY3.

Received: November 1, 2012
Revised: November 23, 2012
Published online: December 19, 2012

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