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# Organic light-emitting diode-based plausibly physiologically-friendly low color-temperature night light

Jwo-Huei Jou [a,*], Ming-Chun Tang [a], Pin-Chu Chen [a], Yi-Shan Wang [a,d], Shih-Ming Shen [a], Bo-Ru Chen [a], Chun-Hao Lin [a], Wei-Ben Wang [a], Szu-Hao Chen [a,d], Chien-Tien Chen [b,*], Fang-Yuan Tsai [b], Ching-Wu Wang [c], Chien-Chih Chen [d], Ching-Chiun Wang [d]

[a] Department of Materials Science and Engineering, National Tsing Hua University, Hsin-Chu 30013, Taiwan, ROC
[b] Department of Chemistry, National Tsing Hua University, Hsin-Chu 30013, Taiwan, ROC
[c] Graduate Institute of Opto-Mechatronics, National Chung Cheng University, Chia-Yi 62102, Taiwan, ROC
[d] Mechanical and Systems Research Laboratories, Industrial Technology Research Institute, Hsinchu 31040, Taiwan, ROC

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ABSTRACT

Light sources with lower color temperature (CT) show markedly less suppression effect on the secretion of melatonin, an oncostatic hormone. Light sources with higher color rendering index (CRI) provide better visual comfort. In this report, we demonstrate the design and fabrication of low CT, high CRI fluorescent organic light-emitting diode (OLED) with five-band emitting from a single emissive layer. The best performed device exhibits a CT of 1773 K, much lower than that of candles (1800–2000 K) or incandescent bulbs (2000–2500 K), 87 CRI, a beyond theoretical limit external quantum efficiency (EQE) 6.4%, and 11.9 lm/W at 100 cd/m$^2$. One major reason for having the ultra-low CT and relative high CRI may be attributed to the significantly intensive deep red emission. The comparatively high efficacy and EQE may be attributed to the employment of a smooth stepwise energy-level structure, enabling low injection barriers and balance carrier injection.



## 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 achieved power efficiency near that of fluorescent tubes. However, investigators have rarely focused on developing physiologically-friendly light sources, especially for use at night [7–14].

Numerous medical studies revealed that frequent exposure to improper light, such as light of high color temperature (CT), or light with short wavelength, at night may increase the risk of cancer types, including cancer of the breast, colorectal, prostate, etc. This is because short wavelength light, or light with high CT, has the effect of markedly high suppressing effect on the secretion of oncostatic hormone, melatonin (MLT) [10–12,15,16]. For examples, at a fixed illuminance, such as 100 lux (lm/m$^2$), the suppression rate of a white LED is three times that of an incandescent bulb, of which the CT ranges from 2000 to 2500 K, or five times that of a candle, of which the CT ranges from 1800 to 2000 K [10]. Therefore, to be physiologically-friendly, the CT of a light source must be low; specifically, it would be ideal to have a light source with CT as low as, or even lower than that of a candle.

In this report, we demonstrate the design and fabrication of a low CT, high CRI fluorescent OLED with five bands emitting from a single emissive layer. The five bands, emitting from blue to deep red light, originate from a blue emission host doped with a fluorescent yellow dye and a

* Corresponding authors. Tel.: +886 3 574 2617 (J.-H. Jou), tel.: +886 3 573 3363 (C.-T. Chen).
*E-mail addresses:* jjou@mx.nthu.edu.tw (J.-H. Jou), ctchen@mx.nthu.edu.tw (C.-T. Chen).

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fluorescent red dye. The resultant best performed device exhibits a CT of 1773 K, which is significantly lower than that of incandescent bulbs (2000–2500 K) and even lower than that of candles (1800–2000 K), as shown in Fig. 1 [17–37]. It is noteworthy that almost all reported white OLEDs showed a CT notably greater than 2500 K, while the CTs for warm and cold fluorescent tubes are 2500–3000 and 4000–5000 K, respectively. The device also exhibits a fairly high CRI of 87 between 100 and 1000 cd/m$^2$, by noting that the CRI is 90 for candles and 98 for incandescent bulbs. The device shows a beyond theoretical limit external quantum efficiency (EQE) [38] of 6.4% at 100 cd/m$^2$, and 5.3% at 1000 cd/m$^2$. Its corresponding efficacy is 11.9 lm/W at 100 cd/m$^2$ and 10.1 lm/W at 1000 cd/m$^2$, while 0.1 lm/W for a candle, or 12–15 lm/W for incandescent bulbs. It is believed that this OLED device, which possesses a CT lower than that of a candle and an efficacy comparable to that of incandescent bulbs, and meanwhile a high CRI, may be a good candidate for use as a light source at night to safeguard human health.

The device structure is comprised of a 125 nm indium tin oxide anode layer, a 25 nm poly(3,4-ethylene-dioxythiophene)-poly-(styrenesulfonate) (PEDOT:PSS) hole injectionlayer (HIL) [39–41], a 22 nm single emissive layer, a 32 nm 1,3,5-tris(*N*-phenylbenzimidazol-2-yl)benzene (TPBi) electron-transporting layer (ETL), a 1 nm lithium fluoride (LiF) electron injection layer, and a 150 nm aluminum cathode layer. The single emissive layer is composed of a fluorescent yellow dye, Spiro-fluorene-dibenzosuberan[d](1,4-bis(4-(*N*,*N*-diphenylamine)-phenyl)-quinoxaline) (Spiro-QDPAP) [42], and a fluorescent red dye, ER55, doped into a blue light emitting host. The dye to host weight ratio is fixed at 5% for the yellow dye and varies between 0.1% and 0.7% for the red dye. Three different blue light-emitting host materials are studied for comparison. They are 2,7-bis{2[phenyl(m-tolyl)amino]-9,9-dimethyl-fluorene-7-yl}-9,9-dimethyl-fluorene (MDP3FL), 1-butyl-9,10-naphthalene-anthracene (BANE), and 2-(*N*,*N*-diphenyl-amino)-6-[4-(*N*,*N*-diphenylamine)styryl]naphthalene (DPASN).

The fabrication process initially involved spin-coating an aqueous solution of PEDOT:PSS at 4000 rpm for 20 s to form the 25 nm HIL. The EML and ETL were deposited using the thermal deposition method in a high vacuum chamber ($3 \times 10^{-5}$ Torr) at a rate of 1–2 Å s$^{-1}$. The electron injection layer (LiF) and aluminum cathode were also deposited using the thermal deposition method in a high vacuum chamber ($3 \times 10^{-5}$ Torr) at respective rates of 0.1 and 12 Å s$^{-1}$.

The ITO glass, three host materials, and TPBi studied herein were purchased from Luminescence Technology Corporation. The hole-transport material (PEDOT:PSS) (Clevios P VPAI4083) was purchased from Heraeus Clevios GmbH Corporation. The LiF was purchased from Strem Chemicals Incorporation. The aluminum cathode was purchased from Showa Chemical Industry Company Ltd. The Spiro-QDPAP was synthesized by Chen et al. [42]. The ER55 was purchased from e-Ray Optoelectronics Technology Corporation, Ltd. All the devices were measured in forward direction without the use of any outcoupling techniques.

The device structures and molecular structures of the employed OLED materials are illustrated in Fig. 2. Table 1 presents the effects of the different host materials and different red dye doping concentrations on the resulting CT, CRI, EQE, efficacy and current efficiency at 100 cd/m$^2$ and 1000 cd/m$^2$.

As shown in Table 1, the best performed Device II, using MDP3FL as host, shows a low CT of 1773 K, a high CRI of 87, and a beyond theoretical limit EQE (efficacy and current efficiency) of 6.4% (11.9 lm/W and 11.9 cd/A) at 100 cd/m$^2$. Whilst, for the device using BANE host (Device V), shows a low CT of 1952 K, a high CRI of 84, and an EQE

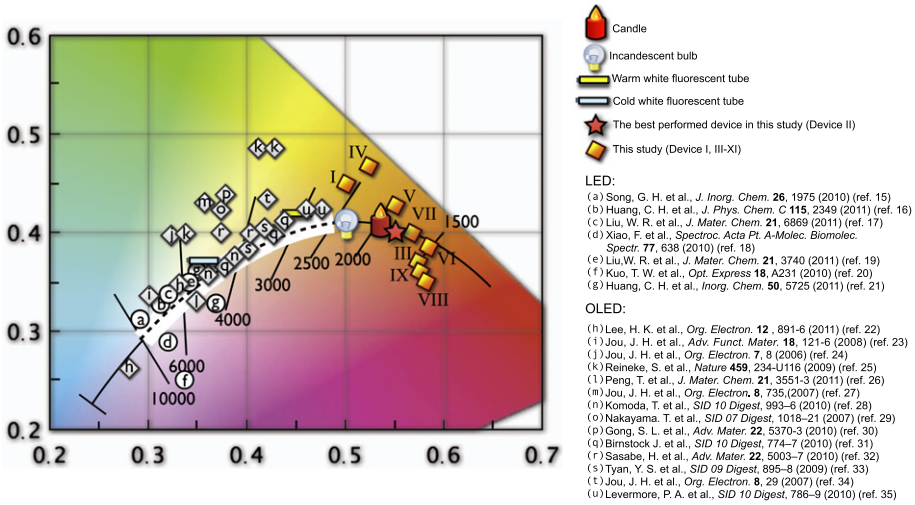


**Fig. 1.** Schematic illustrations of the CIE coordinates of a 2000 K color-temperature (CT) candle, a 2500 K incandescent bulb, a 3000 K warm fluorescent tube, a 4500 K cold fluorescent tube, some prior reported LEDs and OLEDs, and the very-low CT OLED devices fabricated in this study. Nearly all the reported white OLEDs show a CT much greater than 2500 K. Notably, the resultant best performed Device II exhibits a CT of 1773 K, much lower than that of incandescent bulbs (2000–2500 K) and even lower than that of candles (1800–2000 K). The very low CT indicates the OLED device to possess a markedly lower suppression effect on the secretion of the oncostatic hormone, melatonin, and hence, to be a physiologically-friendly light source for use at night to safeguard human health.

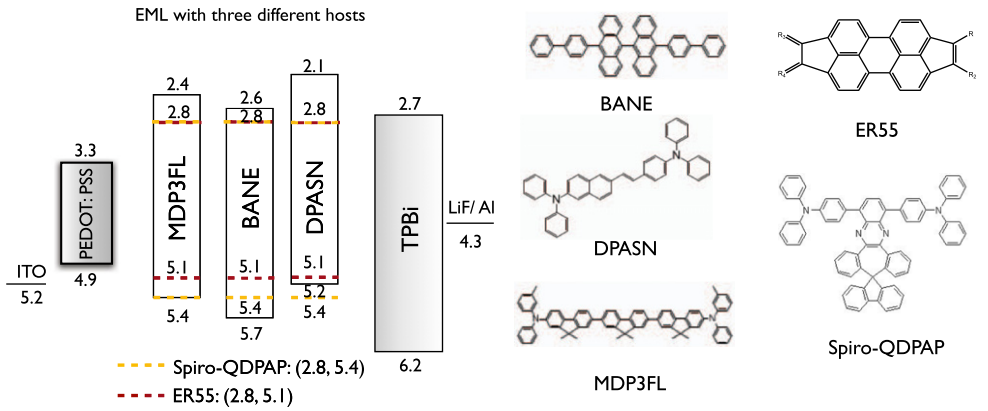


**Fig. 2.** Device structures of the studied low color-temperature (CT) OLEDs and molecular structures of the employed light-emitting guests and hosts. Three different blue light-emitting hosts, MDP3FL, BANE, and DPASN, are respectively investigated in Devices I–III, IV–VI, and VII–XI. Amongst, the devices using the MDP3FL host show a most balance carrier injection, which favors the generation of high energy excitons on host, and hence favors the occurrence of host-to-guest energy transfer, leading to a most effective emission of the guest emitters.

**Table 1**
Effects of host materials and red dye doping concentrations on the resulting CT, CRI, EQE, efficacy and current efficiency of the studied low color-temperature OLEDs at 100 and 1000 cd/m$^2$. Amongst, the best performed Device II that contains the MDP3FL host exhibits an ultra-low CT (1773 K) with a comparatively highest CRI (87) and a beyond theoretical limit EQE (6.4%).

| Device | Host | Weight ratio of red dye (%) | Maximum luminance (cd/m$^2$) | Power efficiency (lm/W) | Current efficiency (cd/A) | External quantum efficiency (%) | Color temperature (K) | Color rendering index | CIE coordinates |
|---|---|---|---|---|---|---|---|---|---|
| *@100/1000 nits* | | | | | | | | | |
| I | MDP3FL | 0.1 | 14,500 | 13.0/8.0 | 12.8/10.1 | 5.8/4.3 | 2504/2552 | 78/77 | (0.50,0.45)/ (0.49,0.45) |
| II | | 0.5 | 18,100 | 11.9/7.6 | 11.9/10.1 | 6.4/5.3 | 1773/1830 | 87/87 | (0.55,0.40)/ (0.54,0.40 |
| III | | 0.7 | 10,300 | 7.1/4.1 | 7.3/5.7 | 4.6/3.6 | 1445/1553 | 81/82 | (0.57,0.37)/ (0.55,0.37) |
| IV | BANE | 0.1 | 31,100 | 8.7/5.9 | 11.4/10.2 | 4.5/4.0 | 2450/2500 | 63/63 | (0.52,0.47)/ (0.51, 0.47) |
| V | | 0.5 | 18,100 | 5.9/3.9 | 8.9/8.0 | 4.3/3.8 | 1952/1980 | 84/84 | (0.55, 0.43)/ (0.54, 0.42) |
| VI | | 0.7 | 16,400 | 3.6/2.5 | 5.3/4.9 | 3.1/2.7 | 1447/1631 | 78/83 | (0.59,0.39)/ (0.57,0.40) |
| VII | DPASN | 0.1 | 5800 | 1.1/0.8 | 2.0/1.6 | 1.1/0.8 | 1690/1906 | 82/84 | (0.56, 0.40)/ (0.52, 0.40) |
| VIII | | 0.5 | 4300 | 1.2/1.0 | 1.7/1.5 | 1.2/1.0 | 1265/1387 | –/76 | (0.59,0.35)/ (0.56, 0.35) |
| IX | | 0.7 | 4400 | 0.9/0.8 | 1.0/0.6 | 0.9/0.8 | 1356/1484 | 79/79 | (0.58,0.36)/ (0.56,0.36) |

The weight ratio of yellow dye is fixed at 5% in every device.

(efficacy and current efficiency) of 4.3% (5.9 lm/W and 8.9 cd/A) at 100 cd/m$^2$. Device V exhibits a fairly high CRI and a fairly low CT, but has the relatively low efficiency. For the device using DPASN host (Device VII), shows a low CT of 1690 K, a high CRI of 82, and an EQE (efficacy and current efficiency) of 1.1% (1.1 lm/W and 2.0 cd/A) at 100 cd/m$^2$. Device VII exhibits an extremely low CT and a fairly high CRI, but has comparatively low efficiency.

The reason why Device II shows a relatively low CT may be attributed to its strongest red emission occurring at above 610 nm. The strong red emission may, consequently, be attributed to the device architecture, having the most balanced carrier injection, wherein the energy barriers for hole and electron to inject into the MDP3FL host are 0.5 eV and 0.3 eV, while 0.8 eV and 0.1 eV for the BANE host, and 0.3 eV and 0.6 eV for the DPASN host. The balanced injection would, hence, favor the generation of high energy excitons on the host, which would consequently favor the occurrence of host-to-guest-energy transfer, leading to effective emission of the guest emitters [43–53].

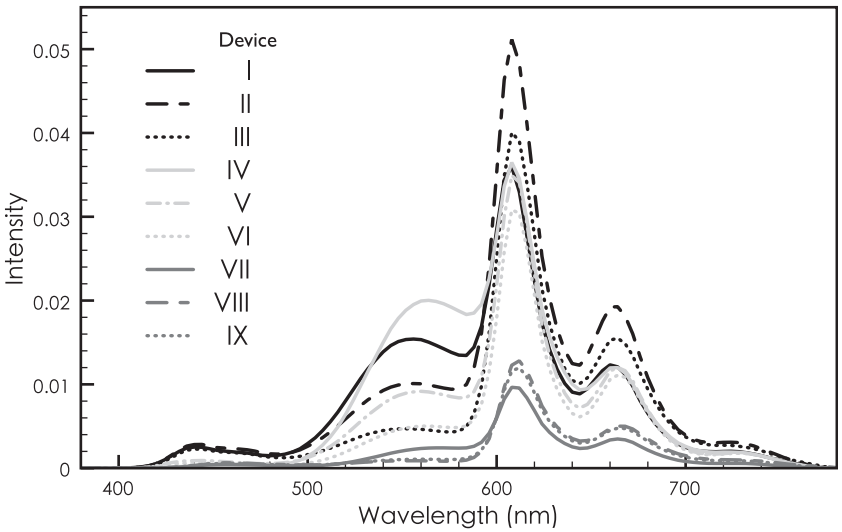


**Fig. 3.** Comparison of the electroluminance spectra of all the 9 low CT OLEDs based on the same current density 10 mA/cm$^2$. All the devices exhibit five bands, ranging from blue to deep-red emission, forming the basis of a high color-rendering index (CRI). Device II shows a strongest red emission at and above 610 nm, in favor the formation of light source with a relatively low CT.

The reason why all the studied devices exhibit fairly high CRI may be attributed to the presence of five-band from the three light emitters, one blue emission from the host, one yellow emission from the yellow dye, and three red emissions from the red dye.

The reason why Device II exhibits the highest device efficiency may mainly be attributed to its most balanced carrier injection, as described previously. The balance in carrier injection is seemingly more critical than other factors, such as effective host-to-guest-energy transfer and effective carrier confinement [43–53]. Taking the host-to-guest-energy transfer for example, the MDP3FL host, used in Device II, performs comparatively poorer than the respective BANE and DPASN hosts, used in Devices V and VII, as indicated by the lesser overlapping area between the PL spectrum of the MDP3FL host, and the UV–Vis spectrum of the yellow dye, Spiro-QDPAP guest, as shown in Fig. 4(a). As shown in Fig. 3, marked blue emission in the EL spectrum provides evidence of less efficient host-to-guest energy transfer for the MDP3FL host. In contrast, only extremely mild blue emission can be observed from the BANE and DPASN blue light-emitting hosts, indicating that both hosts demonstrate more effective energy transfer.

The strong red emission may, consequently, be attributed to the device architecture, having the most balanced carrier injection, wherein the energy barriers for hole and electron to inject into the MDP3FL host are 0.5 eV and 0.3 eV, while 0.8 eV and 0.1 eV for the BANE host, and 0.3 eV and 0.6 eV for the DPASN host.

The DPASN host containing device possesses a highest hole confining barrier, in favor retaining the injected holes in the emissive layer. However, it also possesses a highest barrier for electron to inject from the electron transporting layer to the host leading to a very imbalanced carrier injection, and hence a relatively poorest device efficiency. Furthermore, there is no electron confining barrier between the host and the hole injection layer, PEDOT, due to the fact that PEDOT shows a LUMO value, −3.3 eV, [39–41] much lower than the −2.1 eV of the DPASN host. This would hence worsen the imbalance of the carrier injection, especially at elevated voltages, which may explain why the resulting device (Device VII) showed a more marked current efficiency drop, i.e. 20%, as the luminance increased from 100 to 1000 cd/m$^2$. Whilst, the current efficiency drop were 15% for Device II and 10% for Device V.

Besides host-to-guest-energy transfer, energy transfer from the yellow dye to the red dye is also highly likely, as indicated by the significantly large overlapping area between the PL spectrum of the yellow dye, Spiro-QDPAP guest, and the UV–Vis spectrum of the red dye, ER55 guest, shown in Fig. 4(b). This explains why, in all the device systems examined in this study, the red emission is unanimously predominant among all spectra. However, they all rely fundamentally on a sufficient amount of high energy excitons generated on the employed hosts [54–56]. Hence, the low injection barrier and balanced carrier injection of Device II are indeed in favor of this event.

Fig. 5 shows the resultant EQE, current efficiency, and power efficiency results of the studied OLED devices with low color-temperature. For Devices I–III that compose the MDP3FL host, the respective EQEs are 5.8% or 4.3%, 6.4% or 5.3%, and 4.6% or 3.6%, current efficiencies 12.8 or 10.1, 11.9 or 10.1, and 7.3 or 5.7 cd/A, and power efficiencies 13.0 or 8.0, 11.9 or 7.6, and 7.1 or 4.1 lm/W at 100 or 1000 cd/m$^2$.

For those with the host of BANE (Devices IV–VI), the respective EQEs are 4.5% or 4.0%, 4.3% or 3.8%, and 3.1% or 2.7%, current efficiencies 11.4 or 10.0, 8.9 or 8.0, and 5.3 or 4.9 cd/A, and power efficiencies of 8.7 or 5.9, 5.9 or 3.9, and 3.6 or 2.5 lm/W at 100 or 1000 cd/m$^2$.

As the DPASN host is employed (Devices VII–IX), the respective EQEs are 1.1% or 0.8%, 1.2% or 1.0%, and 0.9%

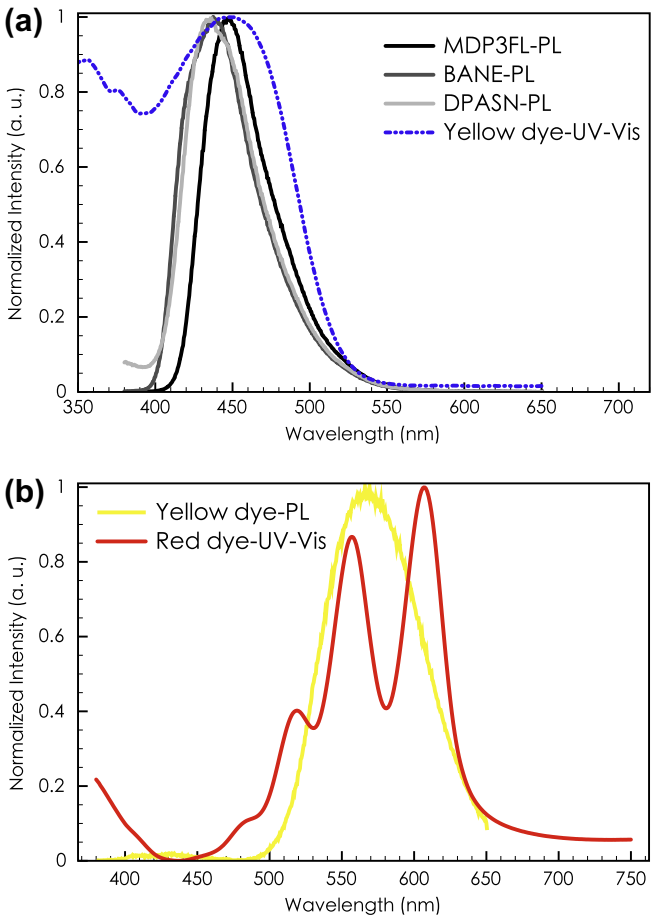


**Fig. 4.** (a) The photoluminescence (PL) spectra of the three blue light-emitting hosts, MDP3FL, BANE, and DPASN, overlapping differently with the UV–Vis spectrum of the yellow dye, Spiro-QDPAP. Lesser overlapping between the PL spectrum of the MDP3FL host and the UV–Vis spectrum of the yellow dye indicates a comparatively poorer host-to-guest energy transfer. (b) The PL spectrum of the yellow dye, significantly overlapping with the UV–Vis spectrum of the red dye, ER55, indicating a high likelihood of effective guest-to-guest energy transfer.

or 0.8%, current efficiencies 2.0 or 1.6, 1.7 or 1.5, and 1.0 or 0.6 cd/A, and power efficiencies 1.1 or 0.8, 1.2 or 1.0, and 0.9 or 0.8 lm/W at 100 or 1000 cd/m$^2$.

By comparing Devices II and VIII, the majority of excitons, at low bias, would form on the dopant because the barrier for hole to enter into the host BANE is relatively higher. The resulting low energy excitons, formed on the dopant, would be more difficult to trigger its own high energy emission. At high bias, holes with higher energy could much easily conquer the injection barrier and increasing excitons could hence form directly on the host. The resulting high energy excitons could, in contrast, easily trigger the high energy emission of the dopant. Since both the host and guest are used to generate excitons at high bias, the increasing excitons may explain why the current efficiency rolls up accordingly. The resulting high population of excitons would increase the likelihood of triplet–triplet annihilation. Since a proportion of the annihilation would subsequently generate singlet excitons to produce additional light, this may explain why a beyond theoretical limit EQE is observed, as also reported in the literature [57].

The reason for Device II showing a beyond theoretical limit EQE, that is, 5% for fluorescent OLEDs, may be attributed to the high population of excitons, which increase the likelihood of triplet–triplet annihilation. A proportion of these triplet–triplet annihilations would subsequently generate singlet excitons to produce additional light, as reported in the literature by Jou et al. [57].

## 2. Conclusion

To conclude, we demonstrate in the report the design and fabrication of an OLED-based light source with an ultra-low CT and a relatively high CRI, coupling with a beyond theoretical limit EQE. The best performed device shows a 1773 K CT, 87 CRI, 6.4% EQE, and 11.9 lm/W efficacy simply using fluorescent materials without any light out-coupling technique [27,58]. The ultra-low CT, high CRI, and high efficiency may be attributed to the strongest red emission at and above 610 nm, the presence of five-band from blue to deep-red, and a most balance carrier injection, respectively. Notably, the resulting CT is much

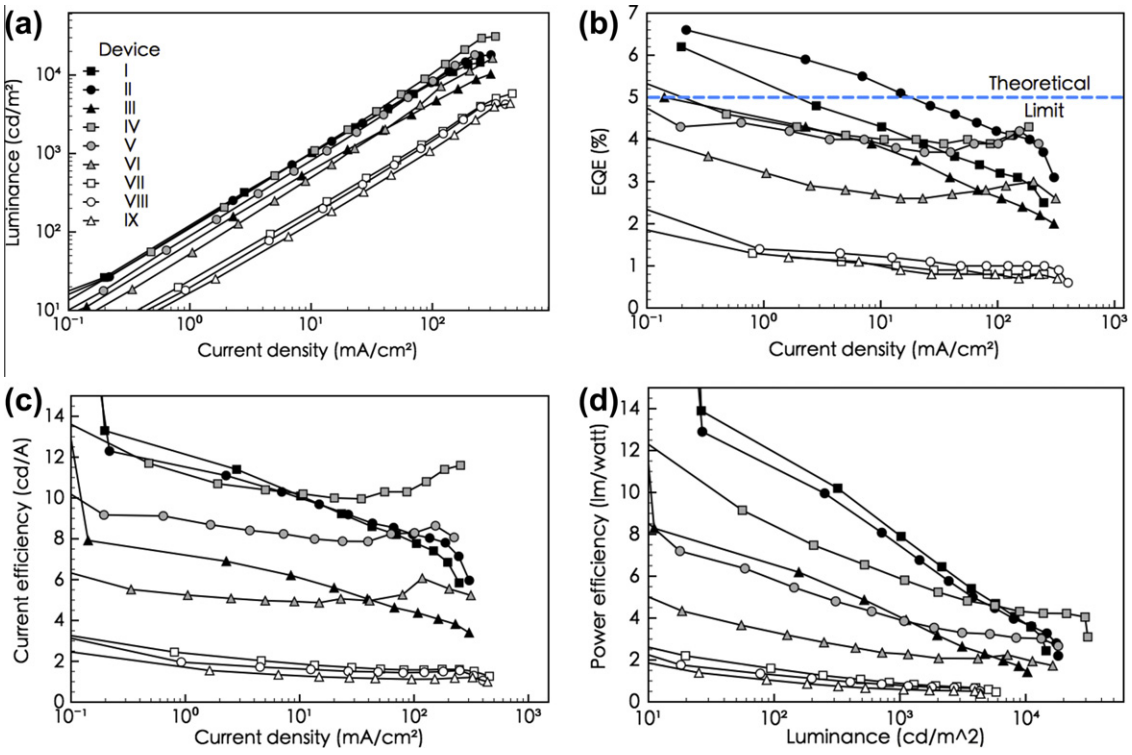


**Fig. 5.** Effects of blue light-emitting hosts and red dye doping concentrations on the (a) luminance, (b) external quantum efficiency, (c) current efficiency and (d) efficacy of the studied low color temperature OLEDs. Notably, the best performed Device II exhibits an EQE much greater the typical 5% theoretical limit between 0.2 and 20 mA/cm$^2$ or 26 and 1880 cd/m$^2$.

lower than that of a candle, indicating which to possibly possess a markedly less suppression effect on the secretion of melatonin, and the resultant high CRI should also warrant a reasonably good visual comfort. These plausibly physiologically- and psychologically-friendly characteristics show OLED to be a very ideal candidate as the lighting source to use at night to safeguard human health.

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