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High-efficiency low color temperature organic light emitting diodes with solution-processed emissive layer

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ABSTRACT

Low color temperature (CT) lighting provides a warm and comfortable atmosphere and shows mild effect on melatonin suppression. A high-efficiency low CT organic light emitting diode can be easily fabricated by spin coating a single white emission layer. The resultant white device shows an external quantum efficiency (EQE) of 22.8% (34.9 lm/W) with CT 2860 K at 100 cd/m^2, while is shown 18.8% (24.5 lm/W) at 1000 cd/m^2. The high efficiency may be attributed to the use of electroluminescence efficient materials and the ambipolar-transport host. Besides, proper device architecture design enables excitons to form on the host and allows effective energy transfer from host to guest or from high triplet guest to low counterparts. By decreasing the doping concentration of blue dye in the white emission layer, the device exhibited an orange emission with a CT of 2280 K. An EQE improvement was observed for the device, whose EQE was 27.4% (38.8 lm/W) at 100 cd/m^2 and 20.4% (24.6 lm/W) at 1000 cd/m^2.

1. Introduction

Color temperature (CT) of a lighting source plays an important role in human physiology and psychology [1–7]. Lighting with low CT, which was generally defined below 5500 K corresponding to CT of pure-white lighting above 5500 K, provides a warm and comfortable atmosphere and helps stabilize autonomic nervous function [8]. Most importantly, it shows a milder suppression effect on the secretion of melatonin (MLT) [9,10]. Being constantly exposed to the light with high color temperature, however, will stimulate the secretion of cortisol that makes people awake and more active [1,3,7] but will markedly suppress the nocturnal secretion of MLT, increasing the risk of being afflicted with cancers, such as breast, colorectal, prostate etc. [4]. Therefore, a proper lighting source with suitable color temperature will be an important issue which most people are unaware of.

In typical lighting options, low CT devices include candles, incandescent bulbs, and warm-white fluorescent lamps. However, the first two aforementioned lighting devices are energy wasting. For example, the respective power efficiency is 0.1 and 15 lm/W. In solid-state lighting, organic light emitting diodes (OLEDs) provide a new alternative, which is energy saving and CT tailorable [11,12]. For example, Leo et al. reported a power efficiency of 38 lm/W with 3180 K at 100 cd/m^2 [13], So et al. mentioned a power efficiency of 40 lm/W with 4970 K [14], and Kido et al. published a finding of 55 lm/W with 5340 K [15]. All these were fabricated via vapor deposition. For solution-processed devices, Tokito et al. reported 16.2 lm/W with 4270 K at 100 cd/m^2 [16], Forrest et al. found a result of 12.2 lm/W with 4750 K [17], and Yang et al. reported a power efficiency of 16 lm/W with 4660 K [18]. Although solution process provides numerous

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advantages, such as high throughput, large-area size and low cost [19], the efficiency of wet-processed devices was apparently lower than that of the vapor deposition processed counterparts. Also, the aforementioned devices have high efficiencies yet their CTs are relatively higher. Therefore, the efficiency of wet-processed OLEDs with low CT needs further improvement. Numerous approaches have been reported to obtain high efficiency, such as electroluminescence (EL) efficient materials, and structure with low carrier injection barrier, effective carrier or exciton confinement, excitons forming on the host, balanced carrier injection, and efficient host-to-guest energy transfer [11,20–29].

This study demonstrates two high-efficiency low CT OLEDs with a solution-processed hole-injection layer (HIL) and an emissive layer (EML). One device showed a power efficiency of 34.9 lm/W (55.9 cd/A) at 100 cd/m^2 or 24.5 lm/W (46.0 cd/A) at 1000 cd/m^2 with white emission of CT ranging between 2860 and 3030 K, while the power efficiency of the other one was 38.8 lm/W (55.9 cd/A) at 100 cd/m^2 or 24.6 lm/W (46.0 cd/A) at 1000 cd/m^2 with orange emission of CT ranging between 2280 and 2320 K.

The high-efficiency low CT white device is comprised of an anode layer of indium tin oxide, a HIL of PEDOT:PSS, an EML containing 0.6 wt.% tris(2-phenylquinoline)iridium(III) [Ir-(2-phq)$_3$] (orange-red dye), 0.2 wt.% bis[5-methyl-7-trifluoromethyl-5*H*-benzo(*c*)(1,5)naphthyridin-6-one]iridium(picolinate) (CF$_3$BNO, green dye), and 14 wt.% bis(3,5-difluoro-2-(2-pyridyl)-phenyl-(2-carboxypyridyl)iridium(III) (FIrpic, blue dye), doped in a 4,4′-bis(carbazol-9-yl)biphenyl (CBP) host, an electron transporting layer (ETL) of 1,3,5-tris(*N*-phenylbenzimidazol-2-yl)benzene (TPBi), an electron injection layer (EIL) of lithium fluoride (LiF) and a cathode layer of aluminum (Al). 3,5-Di(9H-carbazol-9-yl)tetraphenylsilane (SimCP2) and 4,4′,4″-tri(N-carbazolyl)triphenylamine (TCTA) hosts were also used in device fabrication.

Fig. 1 compares the power efficiency of devices devised for this work and other previously reported solution-processed white OLEDs with respect to CT. The best reported power efficiency of solution-processed white OLEDs was 25 lm/W (39 cd/A) [30]. However, its emission is cold-white, i.e. beyond CT 6500 K. On the other hand, for lower CT emissions, such as pure- and warm-white light from 2500 to 6500 K, the best reported efficiency also shown in Fig. 1 was 16.2 lm/W [16]. This study demonstrated a high-efficiency warm-white OLED (marked with II) with a power efficiency of 34.9 lm/W (55.9 cd/A) and CT of 2860 K. In addition to a white device, an orange OLED (marked with VII) with a higher relative power efficiency of 38.8 lm/W (66.5 cd/A) and a lower CT of 2280 K was presented. Its EL spectrum is shown in the inset.

The ambipolar-transport property of the host plays an important role in the resultant current efficiency. Fig. 2 shows the current efficiency of the devices using three different hosts with the same doping concentrations. At all current densities, the CBP-composed device showed the highest efficiency of all. For example, at low current densities, such as 0.1 mA/cm^2 used for displays applications, the CBP-composed device showed an efficiency of 55.9, 32 cd/A for SimCP2, and only 30 cd/A for TCTA. Moreover, at high current densities, such as 10 mA/cm^2 used for illumination applications, the efficiency of the CBP-composed counterpart was 37.4, 26.8 cd/A for SimCP2, and 21.5 cd/A for TCTA. Since CBP and SimCP2 are ambipolar hosts, the better balance between hole and electron injection in the device may explain the better current efficiency of the device. On the other hand, TCTA is a hole-transporting host, which allows more holes to be injected into the EML. This results in a less balanced carrier injection and, in turn, a lower efficiency.

Besides the ambipolar-transporting property, device architecture also plays an important role in the resultant device efficiency. Fig. 3 shows the energy-level diagram of the white devices using three different hosts. To obtain high efficiency, exciton formation on the host is known to be beneficial. For the SimCP2 host with the highest lowest unoccupied molecular orbital (LUMO) of 2.5 eV among the hosts, electrons from the TPBi ETL are favored to hop onto guests with lower LUMO. Besides, the highest occupied molecular orbital (HOMO) of the SimCP2 is 6.1 eV,

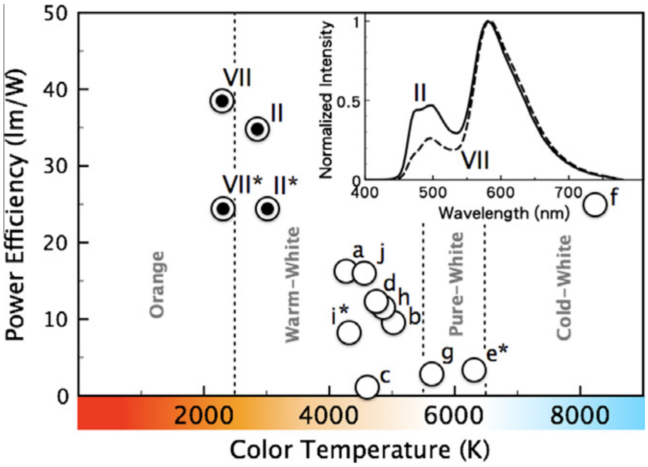

Fig. 1. Power efficiency at 100 cd/m^2 vs. color temperature of the studied devices herein (◉), compared with that of the reported counterparts via solution-process (○). The ones marked with star (∗) show their respective efficiencies at 1000 cd/m^2. Inset shows the EL spectra of our studied OLEDs at 100 cd/m^2 [42].

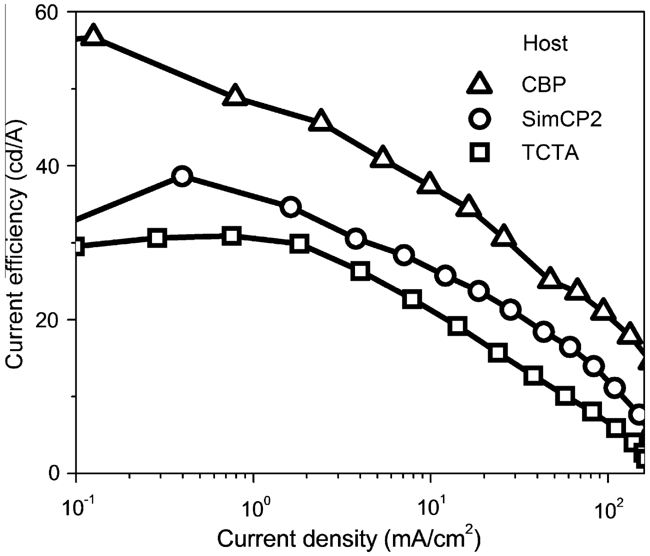

Fig. 2. Current efficiency results of the CBP-composing white device, compared with those of the SimCP2- and TCTA-composed counterparts.

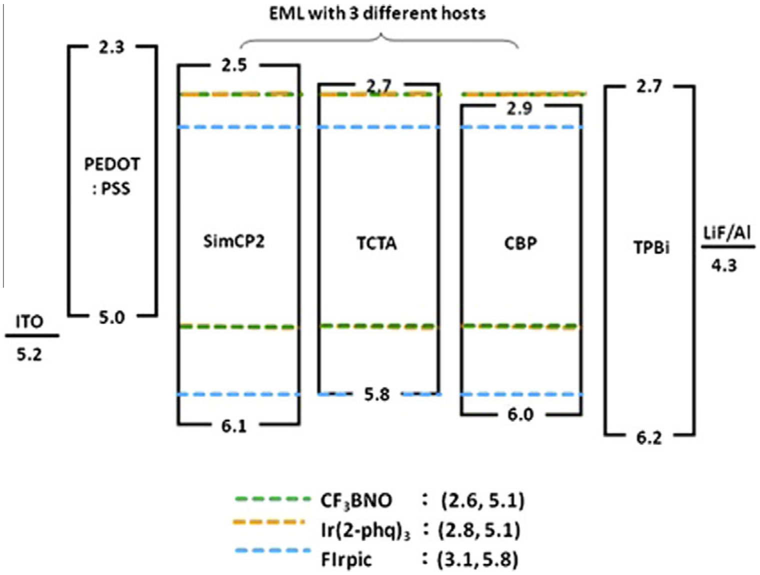

Fig. 3. Energy-level diagram of the hole-injection/emission/electron-transporting structure of the white OLEDs using three different hosts: SimCP2, TCTA, and CBP.

which favors the injection of holes from the PEDOT:PSS HIL to the guests with higher HOMO. This results in the occurrence of exciton quenching on guests, leading to an undesired low efficiency [31–33]. On the other hand, as illustrated in Fig. 3, the CBP host has the lowest LUMO of 2.9 eV among these hosts. Although some electrons may favor injection into the blue dye (−0.4 eV), most electrons inject into the host (−0.2 eV). Since more electrons inject on the host, fewer excitons form on the green and orange-red guests. Thus, the undesired quenching phenomenon is prevented. Hence, an effective host-to-guest energy transfer could be favored by electrons that recombine with holes to form excitons on the host [31–35]. The ambipolar nature and better energy level arrangement of CBP could explain why the CBP-composed device exhibited the best efficiency among all.

Besides the employed hosts, the doping concentrations of the guests play a crucial role in the device efficiency. Table 1 shows the efficiency results by optimizing dye concentrations. By doping 0.1 wt.% green dye with a given concentration of red and blue dyes, the resultant efficiency (Device I) was 27.2 lm/W (43.6 cd/A) at 100 cd/m^2 and 20.4 lm/W (39.0 cd/A) at 1000 cd/m^2. The efficiency was improved to 34.9 lm/W (55.9 cd/A) at 100 cd/m^2 and 24.5 lm/W (46.0 cd/A) at 1000 cd/m^2 as the 0.2 wt.% green dye was doped (Device II). By continuously increasing the doping concentration to 0.3 wt.%, the resultant efficiency of Device III was similar to Device II. However, the

Table 1
Effects of dye-doping concentrations on EL characteristics of the studied CBP-composing device.

Device	Dye concentration (wt.%)			PE (lm/W)	CE (cd/A)	EQE (%)	CT (K)	CIE 1931 (*x*, *y*)
	R	G	B			@ 100/1000 cd/m^2		
I	0.6	0.1	14	27.2/20.4	43.6/39.0	18.0/16.1	2690/2900	(0.47,0.44)/(0.45,0.43)
II	0.6	0.2	14	34.9/24.5	55.9/46.0	22.8/18.8	2860/3030	(0.46,0.44)/(0.45,0.44)
III	0.6	0.3	14	33.5/24.5	53.9/45.6	21.7/18.3	2990/3070	(0.45,0.45)/(0.45,0.45)
IV	0.6	0.4	14	28.0/21.4	53.2/42.1	21.2/16.8	3410/3480	(0.42,0.43)/(0.42,0.43)
V	0.8	0.2	14	31.1/23.7	49.4/43.2	20.0/17.5	2510/2610	(0.48,0.44)/(0.48,0.44)
VI	0.4	0.2	14	23.3/19.2	38.1/36.9	15.3/14.8	3180/3240	(0.44,0.45)/(0.44,0.44)
VII	0.6	0.2	8	38.8/24.6	66.5/49.4	27.4/20.4	2280/2320	(0.50,0.44)/(0.50,0.44)
VIII	0.6	0.2	4	30.8/19.7	50.1/38.5	20.6/15.9	2270/2300	(0.49,0.45)/(0.50,0.44)
IX	0.6	–	14	17.3/12.1	37.7/32.7	16.0/13.9	3350/2920	(0.43,0.44)/(0.45,0.42)
X	0.6	0.2	–	19.2/13.1	40.4/33.7	17.7/14.7	1510/1500	(0.56,0.43)/(0.56,0.43)

efficiency dropped to a lower value as more dye (0.4 wt.%, Device IV) was employed. The high efficiency may be mainly attributed to the use of more efficient EL materials and more effective host-to-guest energy transfer. Moreover, these devices exhibited a warm-white emission in which CT ranged between 2690 and 3070 K.

The device efficiency relied on the concentration of the orange-red dye as well. As seen in Devices II, V and VI, the optimized doping concentration was 0.6 wt.%. All of these devices exhibited warm-white emissions. Table 1also shows the effects of blue dye concentrations on EL characteristics of the studied OLEDs.

As illustrated, by increasing the concentration of blue dye to from 4 to 8 wt.% (Device VII), the resultant device showed an improved power efficiency from 30.8 to 38.8 lm/W at 100 cd/m^2 or from 19.7 to 24.6 lm/W at 1000 cd/m^2. This efficiency improvement may be attributed to the effective host-to-guest energy transfer. However, by continuously increasing the doping concentration to 14 wt.%, the resultant efficiency dropped to 34.9 lm/W at 100 cd/m^2 and 24.5 lm/W at 1000 cd/m^2. The decrease of device efficiency may be mainly resulted from concentration quenching [31–33]. Surprisingly, it did not lower seriously. The plausible reason for the only mildly-dropping efficiency may be due to paths of effective energy transfer from blue guest to host and in turn from host to other two dyes, facilitating efficiency improvement. For efficient devices, the ideal energy transfer should occur from host to guests or from high bandgap guest to low bandgap guest. However, it is possible for the energy to transfer from high guest with high triplet bandgap, e.g. FIrpic, back to host. During this undesired energy transfer, energy dissipation on the host would occur and in turn lower device efficiency. As seen in Table 1, Device VII exhibits the highest efficiency among all the white devices. One of the important reasons for the best efficiency result may be the effective energy transfer from host to guest, e.g. CBP host to CF$_3$BNO green and Ir(2-phq)$_3$ orange-red dyes, and from high triplet guest to low counterparts, e.g. FIrpic blue to green, blue to orange-red and green to orange-red, as shown in Fig. 4. Interestingly, although energy transfer from blue guest back to host happened, device efficiency was not damaged. The back transferred energy could still deliver to green and orange-red dyes, facilitating the achievement of high efficiency.

However, device efficiency was lowered since some routes of effective energy transfer were absent due to no high triplet materials in the EML. As also seen in Table 1,

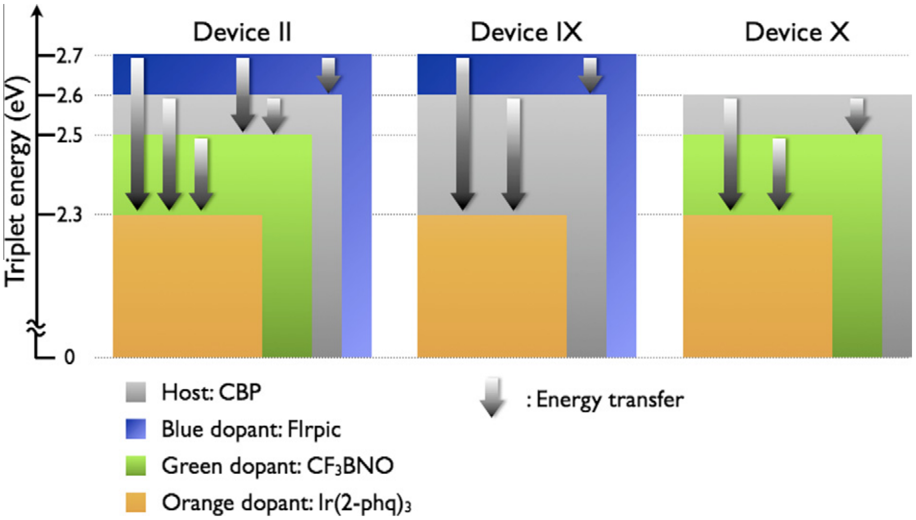

Fig. 4. Proposed energy transfer mechanisms in the white OLED of CBP host and red, green, blue dopants. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Device IX, when compared with Device II, showed a lower power efficiency of 17.3 lm/W (37.7 cd/A) at 100 cd/m^2 or 12.1 lm/W (32.7 cd/A) at 1000 cd/m^2 without the use of green dye. The poorer efficiency may be due to two reasons. One reason may be the lack of effective energy transfer routes from host to green, blue to green, and green to orange-red. The other reason may be the lack of EL efficient green dye, which generally shows a higher relative efficiency than that of both the orange-red and blue dyes. Similarly, as also seen in Table 1, Device X showed a relatively lower power efficiency of 19.2 lm/W (40.4 cd/A) at 100 cd/m^2 or 13.1 lm/W (33.7 cd/A) at 1000 cd/m^2 without the use of blue dye. The low efficiency may be attributed to the lack of effective energy transfer paths from the blue guest to the other two dyes. As one of the EL efficient dyes was absent, these efficiency results may explain why the device showed a decreased efficiency.

In conclusion, this work has demonstrated two high efficiency low CT OLEDs with solution-processed HIL and EML. One device shows a power-efficiency of 34.9 lm/W (55.9 cd/A) at 100 cd/m^2 or 24.5 lm/W (46.0 cd/A) at 1000 cd/m^2 with white emission of CT ranging between 2860 and 3030 K. However, the other one shows 38.8 lm/W (55.9 cd/A) at 100 cd/m^2 or 24.6 lm/W (46.0 cd/A) at 1000 cd/m^2 with orange emission and CT ranging between 2280 and 2320 K. The latter device, which shows lower color temperature with chromaticity near the daylight locus, may be more suitable for use at night due to its presumably lesser suppression effect on melatonin secretion. The high efficiency may be attributed to the use of EL efficient materials, enable excitons to form on the host, balanced carrier injection, and efficient energy transfer from host to guest or from high triplet guest to low counterparts.

2. Experimental

2.1. Materials

Two host materials, 4,4′-bis(carbazol-9-yl) biphenyl (CBP) [36–38] and 4,4′,4″-tris(carbazol-9-yl) triphenylamine (TCTA) [39,40], studied herein were purchased from Luminescence Technology Corporation. The third one, 3,5-di(9H-carbazol-9-yl) tetraphenylsilane (SimCP2) was synthesized by Chen et al. [41]. The two guest materials, bis(3,5-difluoro-2-(2-pyridyl)-phenyl-(2-carboxypyridyl) iridium (III) (FIrpic) and orange-red dye (2-phenylquinoline) iridium(III) [Ir-(2-phq)$_3$], were also purchased from Luminescence Technology Corporation, while the counterpart, bis[5-methyl-7-trifluoromethyl-5*H*-benzo(*c*)(1,5)-naphthyridin-6-one]iridium(picolinate) (CF$_3$BNO), was synthesized by Chin et al. [33]. The hole-transport material poly(3,4-ethylenedioxythiophene)-poly(styrene sulfonate) (PEDOT:PSS) (Clevios P VPAI4083) was purchased from Heraeus Clevios GmbH Corporation.

2.2. Device fabrication

The first step in the fabrication process included a spin-coat of an aqueous solution of PEDOT:PSS at 4000 rpm for 20 s to form the 40 nm HIL on a pre-cleaned indium tin oxide anode. Second, an emissive layer (EML) was deposited by spin coating. The desired solutions were prepared by dissolving the corresponding host and guest molecules in toluene at 45 °C for 30 min with stirring. The employed well-mixed solution was then spin-coated at 2500 rpm for 20 s to form the 35 nm EML under nitrogen. Finally, the electron-transport layer (ETL) (32 nm), the electron-injection layer (HIL) lithium fluoride (0.7 nm), and the aluminum cathode (150 nm) were then deposited at 1×10^{-5} torr.

2.3. Performance measurement of the white OLEDs

The luminance and Commission Internationale de L'Eclairage (CIE) of the resultant white OLEDs were measured by using a Minolta CS-100 luminance-meter. A Keithley 2400 electrometer was used to measure the current–voltage (I–V) characteristics. The emission area of all the resultant devices was 25 mm^2 and only the forward direction luminance was measured.

Acknowledgments

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