Research Article

Organic Light-Emitting Diode with Color Tunable between Bluish-White Daylight and Orange-White Dusk Hue

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The varying color of sunlight diurnally exhibits an important effect on circadian rhythm of living organisms. The bluish-white daylight that is suitable for work shows a color temperature as high as 9,000 K, while the homey orange-white dusk hue is as low as 2,000 K. We demonstrate in this report the feasibility of using organic light-emitting diode (OLED) technology to fabricate sunlight-style illumination with a very wide color temperature range. The color temperature can be tuned from 2,300 K to 9,300 K, for example, by changing the applied voltage from 3 to 11 V for the device composing red and yellow emitters in the first emissive layer and blue emitter in the second. Unlike the prior arts, the color-temperature span can be made much wider without any additional carrier modulation layer, which should enable a more cost effective fabrication. For example, the color-temperature span is 7,000 K for the above case, while it is 1,700 K upon the incorporation of a nanoscale hole modulation layer in between the two emissive layers. The reason why the present device can effectively regulate the shifting of recombination zone is because the first emissive layer itself possesses an effective hole modulation barrier of 0.2 eV. This also explains why the incorporation of an extra hole modulation layer with a 0.7 eV barrier did not help extend the desirable color-temperature span since excessive holes may be blocked.

1. Introduction

Color temperature of light plays a crucial role on human physiology and psychology [1–9]. Bright daylight or high color temperature intensive artificial light, such as cold fluorescent tubes or the latest white LED lamps, stimulates the secretion of cortisol, a hormone that keeps people awake and active [1–3]. Numerous medical studies revealed that frequent exposure to high color temperature light also markedly suppresses the nocturnal secretion of oncostatic melatonin, increasing the risk of breast, colorectal, and prostate cancers [4–7].

Moreover, current lighting sources provide only a fixed color temperature, seriously mismatching what one truly needs from the standpoint of circadian rhythm; that is, circadian rhythm can be entrained by bright light with high color temperature and melatonin generation can be triggered at dark night. Devising a light source with color temperature tunability would hence be highly valuable. However, little

attention had been paid to this until 2009. The first sunlight-style color temperature tunable OLED was reported in 2009, which yielded a wide color-temperature span, fully covering that of the entire daylight locus [10]. However, the corresponding power efficiency was low because of the use of purely fluorescent emitters. Although the efficiency has been much improved as electroluminescence effective phosphorescent emitters were employed, the color rendering index was low [11]. To provide visual comfort, high or very-high color rendering index is required, which can be realized by employing an effective carrier modulation layer (CML) [12]. A high triplet energy CML may effectively regulate the entering carriers into the available wider recombination zones and results in a wide color-temperature span with desirable electroluminescence spectrum [13, 14]. In past decade, several researchers have reported the chromaticity tunable OLEDs using various types of carrier modulation layers. For example, in 2002, Forrest's group has reported that the CIE color coordinates of the OLED emission can be tuned over a

wide range by inserting a 5 nm exciton blocking layer, BCP, between the emissive layers [15]. Chen et al. have reported the emission color of hybrid white OLED can also be tuned by changing the bipolar CML, CBP, thickness from 2 to 8 nm [16]. Recently, our group had also demonstrated the feasibility of using OLED lighting technology to fabricate light sources with low color temperature as well as chromaticity tunable between that of dusk hue and candle-light [17, 18]. The challenge has now become how to design and fabricate a cost effective lighting device with a high color rendering index along with a color temperature tunable character, and daylight chromaticity is essential, especially considering its strong effect on human physiology and psychology [19–22].

We demonstrate, in this report, the feasibility of using OLED technology to fabricate sunlight-style illumination with a very wide color temperature range and high color rendering index (up to 84), without employing a CML. The resulting color temperature is tunable from 2,300 K to 9,300 K, covering that of entire daylight chromaticities. The CIE coordinates of device, composing orange and yellow emitters in the first emissive layer and blue emitter in the second, can simply tune from (0.51, 0.40) to (0.27, 0.31) by changing the applied voltage from 3 to 11 V. The wide color-temperature range may be attributed to the fact that the recombination zone therein can easily be shifted along the different emissive zones from the first to the second layer via voltage control is because the first emissive layer itself possesses an effective hole modulation function for having a 0.2 eV hole injection barrier between the hole transporting layer and the host.

2. Experimental

2.1. Device Fabrication. Figure 1(a) shows the device architectures of the studied sunlight-style OLED devices without any CML. We fabricated the color temperature tunable OLED devices by using three blackbody radiation complementary emitters, that is, a red light-emitting iridium complex with heterocyclic ligand (WPRD931, a proprietary material from Wan Hsiang OLED Ltd.), a yellow light-emitting dye, 5,6,11,12-tetra-phenylnaphthacene (rubrene), and a blue light-emitting dye, EB515 (a proprietary blue light-emitting material from e-Ray Optoelectronics Technology Co. Ltd.), dispersed in two different emissive layers (EMLs). As shown in Figure 1, the first EML (12 nm) for yielding an orange emission was obtained by doping 2 to 4 wt% rubrene and 0.3 to 0.5 wt% of the red dye of WPRD931 in mixed hosts of bis (2-methyl-8-quinolinolato)(4-phenylphenolato)aluminum (BALq) and *N,N'*-Di(1-naphthyl)-*N,N'*-diphenyl-(1,1'-biphenyl)-4,4'-diamine (NPB) with a ratio of 4 : 1, and the second EML (28 nm) was designated to yield a blue emission, which was obtained by doping 10 to 15 wt% EB515 in a host of aryl substituted anthracene derivative (EB43, a proprietary blue light-emitting host from e-Ray Optoelectronics Technology Co. Ltd.). The devices comprised of 5 nm 1,4,5,8,9,11-hexaazatriphenylene-hexanitride (HAT-CN) carrier generation layer (CGL), a 36 nm di-[4-(*N,N*-ditolyl-amino)-phenyl]cyclohexane (TAPC) hole transporting layer (HTL), a 32 nm BmPyPb electron transporting layer (ETL), a 1 nm

lithium fluoride (LiF) electron injection layer, and a 150 nm aluminum cathode layer. All fabricated devices, except Device II-1 and Device II-2, have an additional carrier modulating layer, 1,3,5-tris(*N*-phenylbenzimidazol-2-yl)benzene (TPBi), in between the orange and blue EMLs.

2.2. Characterization. The current-voltage-luminance (*I-V-L*) characteristics of the resulting phosphorescent yellow OLEDs were measured using a Keithley 2400 electrometer together with a Minolta CS-100 luminance meter. Electroluminescence (EL) spectrum and CIE color coordinates were obtained by using a PR655 SpectraScan spectroradiometer. The emission area of all the resultant devices was 9 mm² and only the luminance in the forward direction was measured.

3. Result and Discussion

The OLEDs with a sunlight-style chromaticity were obtained by employing three sunlight chromaticity complementary emitters via device engineering to adjust the relative emissive intensity of the two EMLs so that the resultant emission would fall at or nearby the daylight locus. The three emitters employed had indeed enabled the generation of the desirable sunlight-style chromaticity with a color temperature ranging at least from 2,300 K to 9,300 K. In the present device system, the relative emissive intensity of the two EMLs was achieved simply by adjusting the doping concentration of the yellow emitter in the first EML and the blue emitter in the second EML.

Table 1 summarizes the effect of emitters doping concentration and CML thickness on the resultant color-temperature span. Without the use of CML (Device I-1), the device with a 2 wt% yellow dopant and a 0.3 wt% red dopant in the first EML and 10 wt% blue dopant in the second EML exhibited a color temperature ranging between 2,460 K and 9,340 K. By increasing the red dopant from 0.3 to 0.5 wt% (Device I-2), the orange red emission became dominant, relatively, with a color temperature varying between 2,170 K and 8,990 K. Whilst by increasing the yellow-dopant from 2 to 4 wt% and blue dopant from 10 to 15 wt% (Device I-3), the entire emission slightly shifted toward the bluer side, and color temperature covered the entire day-light locus. This indicates the dopant concentration has played a significant role in obtaining the broader color-temperature span. As a 3 nm CML was inserted in between the orange red and blue EMLs (Device II-1), the blue emission became dominant with a color temperature varying between 2,900 K and 4,890 K.

Notably, the corresponding color-temperature span became markedly smaller, however, as the thickness of the CML was increased to 5 nm (Device II-2). The comparatively weaker blue emission and stronger orange-red emission had resulted in a much lower color temperature along with the smaller color-temperature span of 1,700 K. Apparently, the thicker modulation layer had blocked excessive holes from entering the blue-emissive zone, leading to a blue-less emission. In contrast, more holes would hence be retained in the orange red emissive zone.

With the above-mentioned device architecture, the sunlight-style OLED, Device I-3, exhibited an emission track

FIGURE 1: Schematic illustrations of the device structure, in terms of energy-levels, of the color temperature tunable OLED devices composing three black body radiation complementary emitters, namely, red, yellow, and blue, dispersed in two emissive layers (EMLs) (a) without any hole modulation layer, and (b) with an extra nanoscale hole modulating layer with an energy barrier of 0.7 eV at the interface between the orange and blue EMLs. Nevertheless, there still exists a 0.2 eV hole injection barrier in Device (a) between the hole transporting layer and the host of the first EML, which could hence effectively regulate the injection of hole and in turn the shifting of recombination zone.

closely matching with the day-light locus shown on the CIE chromaticity diagram in Figure 2. Besides having a wide color-temperature span ranging from 2,300 K to 9,300 K, it also emitted a significantly high color rendering index, ranging from 74 to 84.4 for voltage increasing from 3.0 to 11.5 V (Figure 3).

As shown by the electroluminescent spectra in Figure 4, the device initially showed a predominantly orange-red emission spectrum at 4.0 V with CIE coordinates of (0.45,

0.39), turning to pure white (0.33, 0.34) at 7.0 V, and bluish white (0.29, 0.32) at 9.5 V. Relative to the blue emission, the rapidly decreasing peak intensity of the green and yellow emissions with respect to the applied voltage explains why the emission is hypsochromically shifted as the operation voltage increased from 6.0 to 9.5 V. The reason why the sunlight-style OLED, Device I-3, could effectively regulate the shifting of recombination zone is because the first emissive layer itself possesses an effective hole modulation barrier of

TABLE I: Effects of dopant concentration and carrier modulating layer thickness on the color temperature span, color temperature, and color rendering index of the sunlight-style OLED devices studied.

Device	Dopant concentration [wt%]			CML thickness (nm)	CT span (K)	CT (K)	CRI	Power efficiency (lm/W)	Current efficiency (cd/A) @100/1,000 cd/m ²	1931 CIE coordinates	Operating voltage (V)	Maximum luminance (cd/m ²)
	Yellow	Red	Blue									
I-1	2	0.3	10	—	2,460–9,340	2,980/4,530	84.3/80.8	4.0/2.7	5.7/5.4	(0.42, 0.38)/(0.35, 0.35)	4.5/6.1	10,800
I-2	2	0.5	10	—	2,170–8,990	2,940/4,410	85.2/82.1	4.0/2.4	5.7/4.7	(0.43, 0.38)/(0.36, 0.35)	4.5/6.2	10,270
I-3	4	0.5	15	—	2,290–9,280	2,980/4,540	80.7/84.4	3.9/3.1	5.7/6.0	(0.42, 0.38)/(0.35, 0.35)	4.6/6.2	10,160
II-1	2	0.3	10	3	2,900–4,890	3,060/3,870	83.6/84.5	4.3/2.9	5.9/6.0	(0.42, 0.38)/(0.38, 0.37)	4.3/6.4	4,990
II-2	2	0.3	10	5	2,570–4,270	2,860/3,380	80.1/82.0	4.5/2.7	6.3/5.7	(0.44, 0.39)/(0.41, 0.38)	4.5/6.6	4,750

FIGURE 2: Chromaticity and color temperature characteristics of the color temperature tunable OLED, which shows a dusk hue-style emission at low applied voltage and bluish white day light-style emission at high voltage. A record high color-temperature span is observed for the device without using any additional CML.

FIGURE 3: For the studied sunlight-style OLED (Device-I-3), its color temperature changes from 2,290 to 9,280 K and color rendering index from 74 to 84.4 as the applied voltage is increased from 3 to 11.5 V.

0.2 eV. As the operation voltage was increased, increasing electrons could transport to the blue emissive zone and in turn it resulted in a higher probability of recombination therein, leading to a bluer emission as observed. This also explains why the incorporation of an extra CML with a 0.7 eV barrier did not help extend the desirable color-temperature span since excessive holes would have been blocked, and the blocking effect had increased markedly as the CML thickness was increased from 3 to 5 nm.

Figure 5 shows the resultant power efficiency of the studied OLED devices. For the desirable sunlight-style OLED, Device I-3, its respective power efficiency was 3.9 and 3.1 lm/W, and current efficiency 5.7 and 6.0 cd/A, at 100 cd/m² and 1,000 cd/m², respectively.

4. Conclusion

To conclude, we demonstrate in this study a CML free, sunlight-style OLED with color tunable between bluish white

FIGURE 4: Electroluminescence spectra of the sunlight-style OLED (Device I-3) at various applied voltages.

FIGURE 5: The resultant power efficiency of the sunlight-style OLED devices studied. For Device I-3 with the desirable color-temperature span and sunlight-style chromaticity, its power efficiency varied from 4.2 to 1.8 lm/W at luminance increasing from 10 to 10,160 cd/m².

daylight and warm dusk hue with a record high color-temperature span of 7,000 K, along with a color rendering index varying from 74 to 84.4. The reason why the device could effectively regulate the shifting of the recombination zone is because the first emissive layer itself possesses an effective hole modulation barrier of 0.2 eV. Unlike the prior arts, the color-temperature span can be made much wider without any additional CML, which should enable a more cost effective fabrication.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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References

- [1] 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," *Journal of Circadian Rhythms*, vol. 5, article 2, 2007.
- [2] F. A. J. L. Scheer, L. J. P. van Doornen, and R. M. Buijs, "Light and diurnal cycle affect human heart rate: possible role for the circadian pacemaker," *Journal of Biological Rhythms*, vol. 14, no. 3, pp. 202–212, 1999.
- [3] W. J. M. van Bommel, "Non-visual biological effect of lighting and the practical meaning for lighting for work," *Applied Ergonomics*, vol. 37, no. 4, pp. 461–466, 2006.
- [4] 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 Research*, vol. 294, no. 2, pp. 333–339, 1984.
- [5] S. W. Lockley, G. C. Brainard, and C. A. Czeisler, "High sensitivity of the human circadian melatonin rhythm to resetting by short wavelength light," *Journal of Clinical Endocrinology and Metabolism*, vol. 88, no. 9, pp. 4502–4505, 2003.
- [6] S. M. Pauley, "Lighting for the human circadian clock: recent research indicates that lighting has become a public health issue," *Medical Hypotheses*, vol. 63, no. 4, pp. 588–596, 2004.
- [7] T. Hästönen, A. Alila-Johansson, S. Mustanoja, and M.-L. Laakso, "Suppression of melatonin by 2000-lux light in humans with closed eyelids," *Biological Psychiatry*, vol. 46, no. 6, pp. 827–831, 1999.
- [8] 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," *Biological Rhythm Research*, vol. 36, no. 4, pp. 287–292, 2005.
- [9] 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 Research & Technology*, vol. 25, no. 2, pp. 71–80, 1993.
- [10] J.-H. Jou, M.-H. Wu, S.-M. Shen et al., "Sunlight-style color-temperature tunable organic light-emitting diode," *Applied Physics Letters*, vol. 95, no. 1, Article ID 013307, 2009.
- [11] J. H. Jou, S. M. Shen, M. H. Wu, S. H. Peng, and H. C. Wang, "Sunlight-style organic light-emitting diodes," *Journal of Photonics for Energy*, vol. 1, no. 1, Article ID 011021, 2011.
- [12] J.-H. Jou, Y.-C. Chou, S.-M. Shen et al., "High-efficiency, very-high color rendering white organic light-emitting diode with a high triplet interlayer," *Journal of Materials Chemistry*, vol. 21, no. 46, pp. 18523–18526, 2011.
- [13] J.-H. Jou, S.-H. Chen, S.-M. Shen et al., "High efficiency low color-temperature organic light-emitting diodes with a blend interlayer," *Journal of Materials Chemistry*, vol. 21, no. 44, pp. 17850–17854, 2011.
- [14] J.-H. Jou, H.-C. Wang, S.-M. Shen et al., "Highly efficient color-temperature tunable organic light-emitting diodes," *Journal of Materials Chemistry*, vol. 22, no. 16, pp. 8117–8120, 2012.
- [15] W. D'Andrade, M. E. Thompson, and S. R. Forrest, "Controlling exciton diffusion in multilayer white phosphorescent organic light emitting devices," *Advanced Materials*, vol. 14, no. 2, pp. 147–151, 2002.
- [16] P. Chen, W. Xie, J. Li et al., "White organic light-emitting devices with a bipolar transport layer between blue fluorescent and orange phosphorescent emitting layers," *Applied Physics Letters*, vol. 91, no. 2, Article ID 023505, 2007.
- [17] J.-H. Jou, P.-W. Chen, Y.-L. Chen et al., "OLEDs with chromaticity tunable between dusk-hue and candle-light," *Organic Electronics*, vol. 14, no. 1, pp. 47–54, 2013.
- [18] J.-H. Jou, C.-Y. Hsieh, J.-R. Tseng et al., "Candle light-style organic light-emitting diodes," *Advanced Functional Materials*, vol. 23, no. 21, pp. 2750–2757, 2013.
- [19] G. Lambert, C. Reid, D. Kaye, G. Jennings, and M. Esler, "Increased suicide rate in the middle-aged and its association with hours of sunlight," *American Journal of Psychiatry*, vol. 160, no. 4, pp. 793–795, 2003.
- [20] I. Knez, "Effects of colour of light on nonvisual psychological processes," *Journal of Environmental Psychology*, vol. 21, no. 2, pp. 201–208, 2001.
- [21] T. Dalglish, K. Rosen, and M. Marks, "Rhythm and blues: the theory and treatment of seasonal affective disorder," *British Journal of Clinical Psychology*, vol. 35, no. 2, pp. 163–182, 1996.
- [22] R. W. Lam, D. F. Kripke, and J. C. Gillin, "Phototherapy for depressive disorders: a review," *Canadian Journal of Psychiatry*, vol. 34, no. 2, pp. 140–147, 1989.