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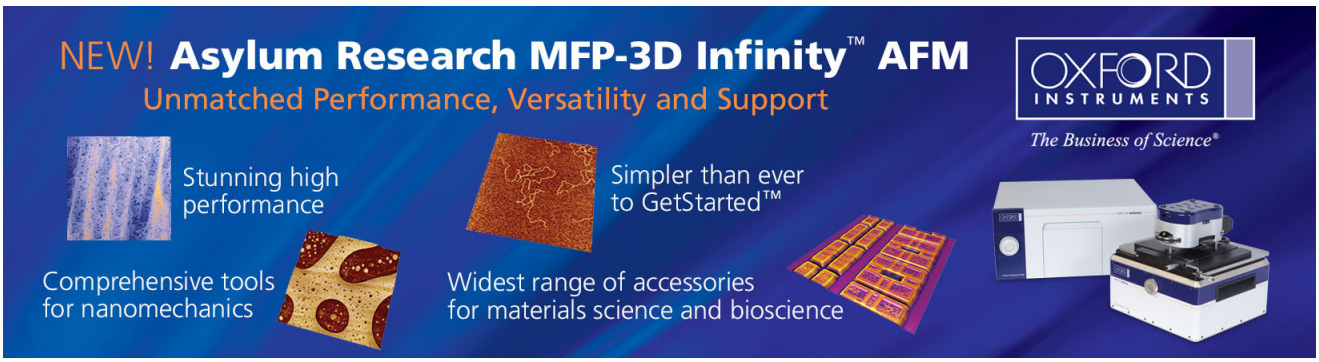

Highly efficient and stable white light organic light-emitting devices

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A highly efficient two-element white light organic light-emitting device with a sky blue dopant BUBD-1 and a yellow dopant TBRb was fabricated. The device achieved an electroluminescence efficiency of 17.1 cd/A and 7.9 lm/W at 20 mA/cm^2. From 112 to 32 010 nits, the CIE$_{x,y}$ coordinates variation was $\Delta(x,y)<\pm(0.01,0.01)$, which is superior to common white light organic light-emitting devices (OLEDs) that tend to change color with drive conditions. Moreover, the device reached a half-decay $t_{1/2}$ lifetime over 40 000 h at an initial luminance of 300 cd/m^2. This result is among the best records of fluorescent white light OLEDs. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2769762]

White light organic light-emitting device (OLED) is a prime focus of OLED research due to its advantages of high resolution and flexible possibility for large-scale production[1] in the applications of full color displays[2] and solid-state lightings.[3] Up to now, enormous efforts have been spent on searching for a low cost and "maskless" method to achieve highly efficient and stable white light. One of the stratagems involves the combination of triple dopants located in one emission zone of the device.[4] This approach can be problematic due to the interdopant energy transfer.[5] In order to achieve a well-balance white color, triple emission layers white light OLEDs have been developed.[6] Nevertheless, undesired chromaticity as well as poor batch-to-batch reproducibility is frequently encountered[7] in such approach, consequently leading to low image-quality displays. For the purpose of lighting, low color rendering index and poor stability also make white light OLEDs far from practical usages. Recently, exciting progresses have been reported in industrial communities.[8–10] One of the best records was reported by Konica Minolta, who has developed a white light OLED with triplet red-green-blue emitters, achieving an extremely high efficiency of 64 lm/W and a half-decay $t_{1/2}$ lifetime of 10 000 h at an initial luminance of 1000 cd/m^2.[8] At the same time, Idemitsu has reported a fluorescent white light OLED with an efficiency of 16.8 lm/W and a long half-decay $t_{1/2}$ lifetime of 30 000 h.[9] Unfortunately, in these reports, full materials structures are not available due to various patents and trade secrets.

Another popular method to achieve white light OLEDs is the two-element stratagem, namely, using a highly efficient sky-blue emitter complemented with a sufficiently broad yellow or orange emitter, to produce white light. This method is attractive due to the lower material cost and simpler fabrication process, in which less dyes and fewer masks are needed. It also provides a better color stability as the aging rate is determined by only two emitters.[11] Therefore, various attentions have been drawn to develop high-efficiency and stable sky-blue emitters, which make great benefits to this kind of white light OLEDs. Up to now, large numbers of phosphorescent and fluorescent sky-blue emitters have been reported.[12,13] A blue phosphorescent OLED with an efficiency of 21 cd/A and a lifetime of 17 500 h was reported.[14] On the other hand, Idemitsu had reported a series of highly efficient fluorescent blue emitters[15] and a two-element white light OLED with an efficiency of 16 cd/A at 100 cd/cm^2 in its patent.[16] However, like most companies, the information of the optimized and energetically matching materials are not disclosed. According to published literature, there was only one report where considerable details have been disclosed with respect to the structures of all electroluminescent materials in the application of white light OLEDs. Ho *et al.* has reported a two-element white light OLEDs with sky-blue dopant 7-DSA-Ph achieving an efficiency of 11 cd/A and CIE$_{x,y}$ coordinates of (x=0.29, y=0.36) at 20 mA/cm^2.[17]

Recently, we have developed a robust sky-blue dopant BUBD-1.[18] In this letter, our objective is to develop a highly efficient and stable two-element white light OLEDs with the benefits of BUBD-1. The device achieved an electroluminescence (EL) efficiency of 17.1 cd/A and 7.9 lm/W at 20 mA/cm^2. Unlike common white light OLEDs, which usually have the deficiency of changing color with drive conditions, the device has a stable color. From 112 to 32 010 nits, the CIE$_{x,y}$ coordinates variation was $\Delta(x,y)<\pm(0.01,0.01)$. Moreover, the white light device reached a half-decay $t_{1/2}$ lifetime over 40 000 h at an initial luminance of 300 cd/m^2. The above device is among the best ever reported for fluorescent emitters based white light OLEDs and represented a significant breakthrough towards the realization of commercial white light OLEDs.

As shown in Fig. 1, three devices have been fabricated. The difference between the devices was that the layer of N,N'-bis-(1-naphthyl)-N,N'-diphenyl,1,1′-biphenyl-4,4′-

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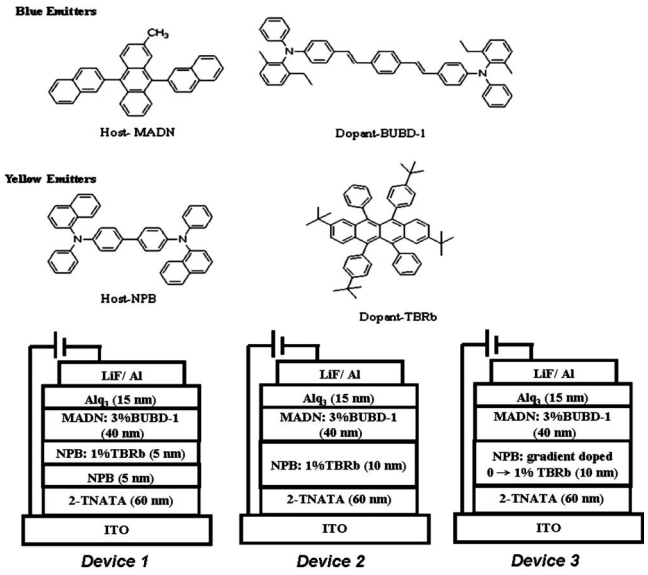

FIG. 1. Configuration and molecular structures of yellow and blue emitters of the devices.

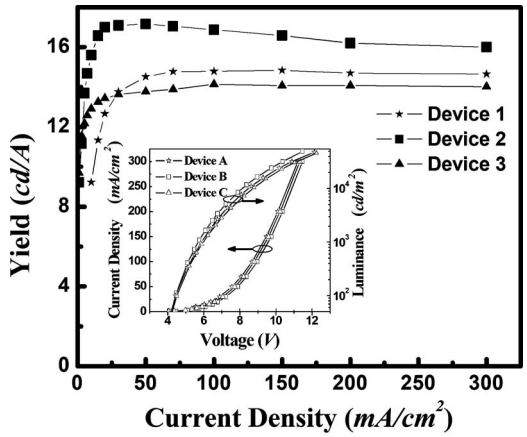

FIG. 2. Dependence of the EL efficiency on the drive current density and inserted is current density–voltage–luminance characteristics for three devices.

diamine (NPB) doped with tetra-(*t*-butyl)-rubrene (TBRb) in devices 2 and 3 was acting as the hole transport layer as well as the yellow emission layer. In device 1, it was only acting as an emission layer, separated by an additional hole transport layer, namely, the nondoped NPB. In device 3, unlike devices 1 and 2, TBRb was doped increasingly from 0% to 1%. The doping ratio of TBRb near the NPB/2-methyl-9,10-di(2-naphthyl)anthracene(MADN) interface was 1%. 4,4′, 4″-tris-[*N*-(2-naphthyl)-*N*-phenylamino]triphenylamine, tris(8-quinolinolato)aluminium (Alq_3) and LiF/Al were used for hole injection, electron transport, and electron injection layer. TBRb is a yellow dopant material which has a higher efficiency than that of rubrene as reported by Wu *et al.*[19] MADN doped with BUBD-1 was used for blue emission layer. Prior to the deposition of organic materials, the indium tin oxide was cleaned with a routine cleaning procedure[20] and pretreated with UV-ozone. The device was fabricated in a standard coater under the base vacuum of about 10^{-6} Torr. The characteristics of current-voltage-luminance of the devices have been measured by PR650 spectrophotometer with a dc source controlled by a connecting computer.

Table I shows a summary of the EL performances of the three devices. Device 2 with a 10 nm doped yellow emission layer achieved a higher efficiency than that of devices 1 and 3. This was probably due to more holes trapped in device 2 and thus improved the carrier balance and increased the overall efficiency. We also noticed that there was essentially no EL color shift with increased driving currents in these devices. For example, in device 2, the $CIE_{x,y}$ color coordinates only shifted from (x=0.285, y=0.406) at 112 nits to (x=0.276, y=0.400) at 32 010 nits with $\Delta(x,y) = \pm(0.009, 0.006)$. Devices 1 and 3 also showed similar behavior except that the overall efficiencies were lower. We attributed the increased device efficiency and stable color to an improved balance between hole and electron currents arriving at the recombination zone, as shown in Fig. 2. It has been reported that doping of hole-trapping materials, such as rubrene, in hole transport layer (HTL) can improve carrier balance.[21] The efficiency of device 2 rises sharply at low current density to the maximum of about 17.1 cd/A at 20 mA/cm^2, then falls down slowly at higher current density. Up to about 200 mA/cm^2, the efficiency is still maintained at about 17 cd/A.

The current density–voltage–luminance characteristics of the three white light OLEDs are also showed in Fig. 2. For device 2, a brightness of 3418 cd/cm^2 was achieved at 20 mA/cm^2. The threshold voltages of the three devices are around 3.0 V, and higher operating voltage was observed in device 2. This phenomenon is consistent with our concept that more holes are trapped in device 2. The influence of hole trapping in device 2 was most significant among the three devices as most TBRb was doped in NPB layer, contributing an enhancement of the yellow emission and the overall efficiency of the device. It also seems that 5 nm doped layer or gradient doped of TBRb in device 1 or 3 were insufficient insofar as to hinder excess holes from achieving an improved hole-electron balance.

TABLE I. EL performances of three devices driven at 20 mA/cm^2.

Device	Cur. Den. (mA/cm^2)	Voltage (V)	Yield (cd/A)	Efficiency (lm/W)	EQE (%)	CIE x	CIE y
	20	6.4	12.7	6.2	4.6	0.3	0.41
1	200	10.1	14.7	4.6	5.6	0.29	0.4
	20	6.8	17.1	7.9	6.4	0.29	0.41
2	200	10.4	16	4.8	6.2	0.28	0.4
	20	6.6	13.4	6.4	5	0.26	0.39
3	200	10.3	14.1	4.3	5.5	0.26	0.38

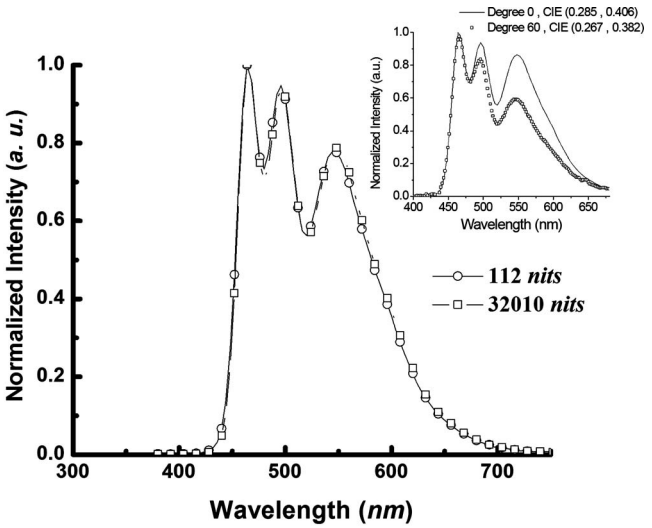

FIG. 3. EL spectrum of device 2 at 1 and 200 mA/cm^2 and EL spectrum of device 2 with different viewing angles.

The EL spectrum of device 2, as depicted in Fig. 3, which covered a wide range of visible region, clearly indicates the emissions of BUBD-1 and TBRb with dominant peaks at 464, 488, and 548 nm, respectively.[18,19] As the figure inserted in Fig. 3 showed, when the viewing angle was varied from 0° to 60° the emission peak of BUBD-1 stayed almost the same while the intensity of the emission peak (548 nm) of TBRb decreased with increased viewing angle. Therefore, the $CIE_{x,y}$ of device 2 changed from (x=0.285, y=0.406) to (x=0.267, y=0.382). We attribute this phenomenon to the optical interference rather than the shifting of the recombination zone which was normally caused by the varying driven current. This is because the device was driven at a fixed current density of 20 mA/cm^2 when varying viewing angle measurements were taken in the atmosphere.

The white light OLEDs also showed exceptionally long half-decay $t_{1/2}$ lifetimes, as shown in Fig. 4. The white light OLEDs were measured at the constant current densities of 20, 30, and 40 mA/cm^2, respectively. Assuming the scalable law of Coulombic degradation of ($L_0^n x t_{1/2}$=const) under accelerated drive conditions[22,23] and by estimation of extrapolated profile, driving at a L_0 value of 300 cd/m^2, the halflife-times of the white light OLEDs were projected to be more than 40 000 h.

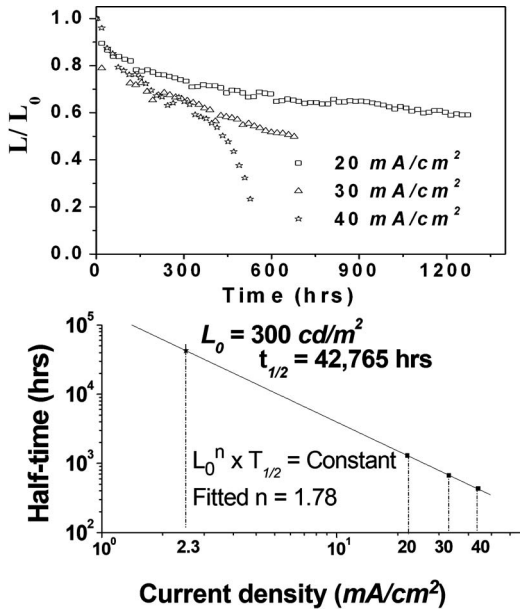

FIG. 4. Device operational stability for device 2.

In summary, we have developed a white light OLED with stable color chromaticity with respect to drive current density. With the employment of sky-blue dopant BUBD-1, we have achieved one of the highest efficiencies and longest operational lifetime white light OLEDs that have been reported in the literature. Based on this work, we believe it will impact positively on the development of both passive and active matrix full color OLEDs in the future.

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