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Low efficiency roll off at high current densities in Ir-complex based electrophosphorescence diode with exciton diffusing and fluorescence compensating layers

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We demonstrate a *fac*-tris(2-phenylpyridine) iridium-based electrophosphorescent organic green-light emitting diode with a considerably reduced current-efficiency roll off at high current density. Such a low roll off of efficiency was achieved by inserting nondoped 4,4′-*N*,*N*′-dicarbazole-biphenyl (CBP) layer and a tris-(8-hydroxy-quinoline) aluminum (Alq_3) layer between the emitting and electron-transporting layers to diffuse excitons from the emitting layer. The Alq_3 layer is found to contribute as a complementary green fluorescent emitter at high current density. Thus, only a small decrease from 20.7 to 16.7 cd/A was detected in current efficiency when the current density increases from 3.9 to 100 mA/cm^2. A high current efficiency of 12.6 cd/A was achieved even at 350 mA/cm^2, indicating that the efficiency roll off was drastically reduced when compared with the device without CBP and Alq_3 layers. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2805740]

Since Tang and Van Slyke demonstrated organic light emitting diode (OLED) based on fluorescent tris-(8-hydroxy-quinoline) aluminum (Alq_3),[1] many progresses have been made to obtain high electroluminescence (EL) performance. The significant improvement in the EL efficiency has been achieved by using electrophosphorescent (EPH) materials, which led to the prospective OLED devices with high internal quantum efficiency of nearly 100%. The typical EPH materials are green-emitting *fac*-tris(2-phenylpyridine) iridium [$Ir(ppy)_3$] and red-emitting platinum octaethylporphyrin (PtOEP), which were firstly used by Baldo *et al.*[2] While triplet emitters have improved the quantum efficiency, however, phosphorescent OLED's suffered from a decrease of efficiency when the current density is increased (called roll off). It has been suggested that triplet-triplet (*T*-*T*) annihilation is mainly responsible for this roll off[3,4] although the other effects such as triplet-charge carrier annihilation, triplet-polaron annihilation and field-assisted dissociation of intermediate state are also responsible.[4,5] This serious disadvantage of high roll-off hinders the prospective application of OLEDs in high-brightness displays, passive matrix-driven displays, solid-state lighting lamp, etc.

Several methods have been proposed to reduce the roll off of efficiency. Recently, He *et al.*[6] and Kang *et al.*[7] proposed $Ir(ppy)_3$-based OLEDs with double emitting layers to confine the triplet exciton, while Cochi *et al.*[8,9] proposed OLEDs with Pt-complex to minimize electric-field-enhanced dissociation of the triplet exciton and to enhance the EL efficiency at high current densities. They tried to confine the triplet excitons in a narrow emitting zone, but the exciton density was increased, leading to drop of the EL efficiency at high current densities. Therefore, a detailed knowledge on the EL quenching at high current densities is necessary to improve the performance of the phosphorescent OLEDs.

In the present work, $Ir(ppy)_3$-based OLED device which shows very low roll off at high current densities was constructed. This was achieved by using an exciton diffusing layer and a fluorescence compensating layer, i.e., we inserted a neat 4,4′-*N*,*N*′-dicarbazole biphenyl (CBP) layer and a Alq_3 layer between $Ir(ppy)_3$-doped CBP emitter layer and the 1,3,5-tris(*N*-phenylbenzimidazol-2-yl)-benzene (TPBi) electron transport layer (see Fig. 1). The nondoped CBP works as an exciton evacuating (i.e., diffusing) layer. By attaching the nondoped CBP layer to the $Ir(ppy)_3$-doped CBP emitter layer, it is expected that excitons can diffuse from the emitting layer to the nondoped CBP layer.[10] As a result, exciton density in the emitting layer is decreased, leading to reduction of the *T*-*T* annihilations in the $Ir(ppy)_3$-doped layer. The Alq_3 layer was intended to store the excitons, which enter

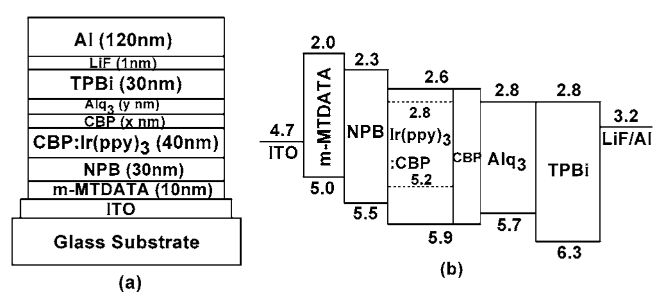

FIG. 1. (a) Device structure of OLED and (b) its proposed HOMO and LUMO energy level diagram.

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into nondoped CBP layer, and to give rise to green emission at 524 nm, resulting in compensation of the quenched green emission at 510 nm from $Ir(ppy)_3$.

By changing the layer thickness of each of the nondoped CBP and Alq_3 layers, we found an $Ir(ppy)_3$-based OLED with very low roll off of current efficiency at high current densities. The best EL performance was obtained from an OLED containing the neat CBP and Alq_3 layers with thickness of 6 and 8 nm, respectively, i.e., very small efficiency roll off at current density range from 1 to 200 mA/cm^2. Furthermore, a current efficiency of 15 cd/A was obtained at 300 mA/cm^2, which is considerably higher than the efficiency of conventional EPH OLEDs.

The fabricated OLED structure is indium tin oxide (ITO)/*m*-MTDATA (10 nm)/NPB (30 nm)/CBP:$Ir(ppy)_3$ (7 wt %, 40 nm)/CBP (x nm)/Alq_3 (y nm)/TPBi (30 nm)/LiF (1 nm)/Al (120 nm) (see Fig. 1). In the parentheses, the layer thickness is indicated. The nondoped CBP layer thickness x was changed from 0 to 20 nm, while the Alq_3 layer thickness y was changed from 0 to 10 nm. The OLED with $x=y=0$, i.e., ITO/*m*-MTDATA (10 nm)/NPB (30 nm)/CBP:$Ir(ppy)_3$ (7 wt %, 40 nm)/TPBi (30 nm)/LiF (1 nm)/Al (120 nm), is called reference device, hereafter. The materials *m*-MTDATA and NPB denote 4,4′,4″-tris[3-methyl-pheny(phenyl)-amino]-triphenyl-amine and 4,4′-bis[*N*-(1-napthyl)-*N*-phenyl-amino]-biphenyl, respectively, which work as the hole-injection layer and hole transporting layer (HTL). The 7 wt % concentration of $Ir(ppy)_3$ in CBP was selected since the high quantum efficiency has been obtained at around 6 wt %.[2] Figure 1(b) shows the energy level diagram of the OLEDs fabricated in this work, where the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) energies are obtained from literatures.[2,11]

The OLED devices were fabricated using precleaned ITO glass (25 Ω/sq) by thermal evaporation at a base pressure of 3×10^{-4} Pa, followed by evaporation of a LiF buffer layer and an Al cathode in the same vacuum run. Deposition rates and thickness of the layers were monitored *in situ* using an oscillating quartz monitor. The evaporating rates were kept at 2–3 Å/s for both organic layers and LiF layer, while 10 Å/s for Al cathode. The EL spectra were measured with a Hitachi F-4500 fluorescence spectrophotometer. The luminance-current-voltage (*L-I-V*) characteristics of OLEDs were measured with an Array Electronic 3645 dc power supply combined with a Photo Research 1980A digital spot photometer. All the measurements were carried out at room temperature under ambient conditions.

The following method was used to find the best combination of the neat CBP and Alq_3 layer thicknesses x and y that gives very low roll off at high current densities. Firstly, the thickness of the neat CBP layer (x) was changed variously at a fixed Alq_3 thickness (y), and then the Alq_3 thickness (y) was changed variously under the determined x value. We repeated this process within the range of $0 < x$ (nm) < 20 and $0 < y$ (nm) < 10. Finally, the OLED with $x=6$ nm and $y=8$ nm was selected as an optimized device that gives the low efficiency roll off at high current densities. Figures 2(a) and 2(b) show the dependences of the current efficiency on current densities for OLEDs with various x and y values, together with the reference device ($x=y=0$). It is seen from these figures that not only high current efficiency

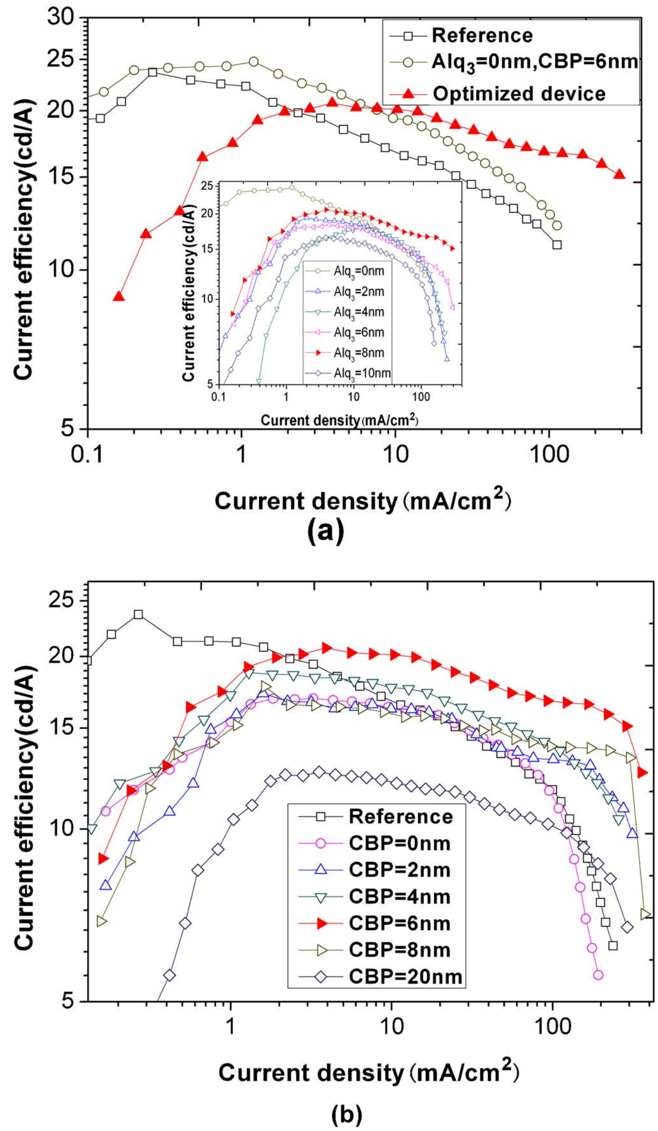

FIG. 2. (Color online) (a) Relation between the current efficiency and current density of OLED devices with $x=6$ and $y=8$ (triangle), $x=6$ and $y=0$ (circle), and $x=y=0$ (square), where x and y mean nondoped CBP and Alq_3 layer thickness (in nanometer units), respectively. Inset: the current density–luminous efficiency characteristics of the devices with various Alq_3 layer thickness and a fixed CBP layer thickness of $x=6$. (b) The current density–efficiency characteristics of the devices with various nondoped CBP layer thickness and a fixed Alq_3 layer thickness of $y=8$, together with a device of $x=6$ and $y=8$ (square).

at high current densities but also low roll off (i.e., much smaller decrease of efficiency with increasing current density above about 10 mA/cm^2) is obtained from the OLED with $x=6$ nm and $y=8$ nm.

A maximum efficiency of 20.7 cd/A is obtained at 3.9 mA/cm^2 in the optimized device ($x=6$ nm, $y=8$ m). As current density increases, the efficiency decreases a slightly and reaches to 16.7 cd/A at 100 mA/cm^2, and then drops to 12.6 cd/A at 350 mA/cm^2. This is quite different from the reference OLED device with $x=y=0$; the efficiency decreases by 50% (i.e., 12 cd/A) at 100 mA/cm^2 from the maximum efficiency of 24 cd/A at 0.3 mA/cm^2 and finally drops to 7 cd/A at 200 mA/cm^2. It is noticed that the insertion of nondoped CBP and Alq_3 layers improves the OLED performance (low roll off and high EL intensity) at high current densities in a range from 10 to 200 mA/cm^2, which is usually employed in the operation in conventional

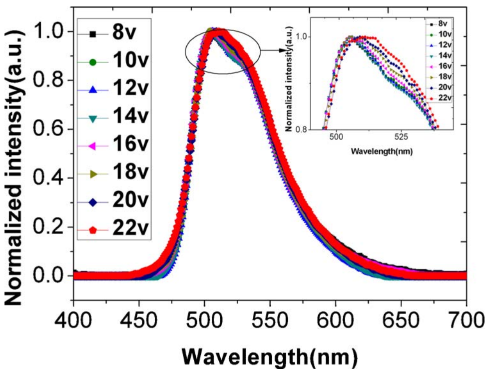

FIG. 3. (Color online) EL spectra of the OLED with nondoped CBP layer thickness of 6 nm and Alq_3 layer thickness of 8 nm at various bias voltages. Inset: enlarged spectra at 490–540 nm.

OLEDs,[3,5] although the current efficiency is much lower in the optimized OLED at low current density below 1 mA/cm^2 than in the reference OLED without nondoped CBP and Alq_3 layers. In fact, regarding the EL luminance, the optimized OLED device ($x=6$, $y=8$) gives 34 000 cd/m^2 at 200 mA/cm^2, while only 14 000 cd/m^2 in the reference device ($x=y=0$) at the same current density. It is also noticed that large thickness of $x>6$ nm and $y>8$ nm leads to high roll off and low EL luminance at high current densities.

As mentioned above, a reduced roll off at high current densities was achieved by introduction of the neat CBP layer. This is understood as follows. The neat CBP layer acts to dissipate excitons in the emitting layer. Strong energy transfer from the CBP host to $Ir(ppy)_3$ dopant, which gives rise to high density excitons at the emitting layer, was avoided. As a result, the *T*-*T* annihilation is reduced. Triplet exciton energy transfer from the emitting layer to the adjacent Alq_3 layer is conceivable because the T_1 state of Alq_3 is much lower than that of $Ir(ppy)_3$.[3] The presence of such an energy transfer is confirmed from Fig. 2(a) where the efficiency is decreased drastically by introduction of Alq_3 at low current density below about 2 mA/cm^2, indicating dissipation of triplet excitons from $Ir(ppy)_3$. The exciton dissipation to Alq_3 layer is favorable to enhance the EL efficiency at high current densities because confinement of high-density excitons in the emitting layer is avoided.

Figure 3 shows the normalized EL spectra of the optimized OLED device ($x=6$ nm, $y=8$ nm) at various bias voltage. It is observed that the 510 nm green $Ir(ppy)_3$ emission peak shifts to low energy with increasing the bias voltage, and emission intensity at about 524 nm enhances. Taking into account that Alq_3 emits EL band with a peak at around 524 nm, it is suggested that the shift is caused by the enhancement of the emission from Alq_3 layer because the recombination zone extends from $Ir(ppy)_3$-doped CBP layer to the Alq_3 layer at high electric field. The green fluorescence from Alq_3 compensates the partial loss of green phosphorescence from $Ir(ppy)_3$ at high electric field, leading to increase the overall EL intensity. Unlike the result of Kang *et al.*,[7] emissions at about 450 nm due to the NPB HTL is not observed even at high voltage (i.e., high current density), as shown in Fig. 3. Kang *et al.* have suggested that the exciton formation in HTL plays an essential role for the efficiency roll off. However, such a suggestion is not applicable for the present OLEDs.

In summary, we achieved a high current efficiency and low efficiency roll off at high current densities using $Ir(ppy)_3$-based OLED with a nondoped CBP layer of thickness 6 nm and a fluorescent Alq_3 layers of thickness 8 nm. Unlike the conventional method of confining the electron-hole recombination zone, the nondoped CBP layer was introduced to diffuse the excess triplet excitons in $Ir(ppy)_3$-doped CBP layer. The green fluorescence from Alq_3 layer compensates the decreased green emission of $Ir(ppy)_3$ at high current density. Both of them contribute to reduce roll off of efficiency. As a result, the maximum current efficiency has just decreased from 20.7 to 16.7 cd/A as the current density has increased from 3.9 to 100 mA/cm^2, i.e., only about 19% efficiency drop was found. It is expected that the OLED layer structure which is proposed in this work is also useful in developing highly efficient OLEDs using other EPH materials. In near future we will confirm it using another phosphorescence material.

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