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Red Phosphorescent Organic Light-Emitting Device Using the Mixed Host of mCP and Bebq₂ in the Emissive Layer

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New red light-emitting phosphorescent devices using the mixed host of N,N'-dicarbazolyl-3,5-benzene [mCP] and bis(10-hydroxy-benzo[h]quinolino)beryllium [Bebq₂] doped with 5 vol.% RP411 (red phosphorescent dye from SFC Co.) in the emissive layer were fabricated and evaluated according to the mixture ratio of x in a host of mCP(1- x) Bebq₂(x).

Among the fabricated devices, the maximum quantum efficiency was obtained in the device with a host of $x = 0.75$ and the highest luminance at a given voltage was obtained in the device with a host of $x = 0.25$. On the other hand, the product of luminance and quantum efficiency showed the best characteristics in the device with a host of mCP(0.5) Bebq₂(0.5). The peak wavelength in the electroluminescence spectra and color coordinates on the Commission Internationale de l'Eclairage (CIE) chart of the fabricated devices with $x \geq 0.25$ in a host of mCP(1- x) Bebq₂(x) were 627 nm and (0.66, 0.34), respectively.

Keywords Bebq₂; efficiency; mCP; mixed host; red phosphorescence

Introduction

A proper combination of host and dopant system in the emissive layer is vital to obtain high performance phosphorescent organic light-emitting diodes (OLEDs). The use of phosphorescent dye as a dopant can greatly improve the quantum efficiency in the OLED because both singlet and triplet excitons can be harvested [1, 2]. The mCP has been widely used as a common host material of three primary color (red, green and blue) emissions in the development of white phosphorescent OLEDs because of its moderate energy gap of 3.6 eV and high triplet energy of 2.9 eV [3, 4]. However, the eligibility of mCP as a red host material is not desirable because the excited energy difference between the host and a light-emitting dye is too large for good energy transfer from host to dye. Therefore, high performance in the electroluminescent characteristics cannot be obtained in the red or white device using a single material of mCP as a host due to low quantum efficiency of red emission. In addition, current density of the device with a host of mCP is relatively low due to hole preferential transport of mCP. The hole and electron mobilities reported in the mCP are 1.2×10^{-4} cm²/Vs and 4.0×10^{-5} cm²/Vs, respectively [5].

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The Bebq₂ has been also used as a host material of red emission in the OLEDs because of its high electron mobility and good energy transfer characteristics [6, 7]. However, current density of the device with a host of Bebq₂ is not high sufficiently due to electron preferential transport of Bebq₂, which gives low luminance characteristics in spite of high quantum efficiency in the fabricated device.

In this study, new red-emitting phosphorescent OLEDs using the mixed host of mCP and Bebq₂ [mCP(1-x)Bebq₂(x)] doped with 5 vol.% dye of RP411 [8] in the emissive layer were fabricated and evaluated. In the experiments, the mixture ratio (x) of Bebq₂ in the host were varied with the values of 0, 0.25, 0.5, 0.75 and 1. Here, x = 0 means the host of mCP only and x = 1 means the host of Bebq₂ only. The proper mixture of mCP and Bebq₂ as a new host structure can improve luminance as well as quantum efficiency due to the good balance of charge transport.

Experimental Procedure

After cleaning the substrate coated with an indium tin oxide (ITO) of 12 Ω/sq on glass, the ITO film was patterned to form an anode by photolithographic technique. To improve the surface morphology of anode, the plasma treatment was executed at 150 W for two minutes under 8 mTorr pressure of O₂/Ar. The plasma treatment before deposition of organic materials is expected to reduce the energy barrier for hole injection from anode and remove the surface contaminants. All organic layers and cathode were deposited by in-situ method under 5×10^{-8} Torr.

As a sequence of process in the fabrication of the devices, the N,N'-diphenyl-N,N'-bis-[4-(phenyl-m-tolyl-amino)-phenyl]-biphenyl-4,4'-diamine (DNTPD) with thickness of 500 Å was firstly deposited as a hole injection layer. And then, a 300 Å -thick film of 1,1-bis-(di-4-polyaminophenyl)cyclohexane (TAPC) was formed as a hole transport layer. In the formation of emissive layer (EML), 300 Å-thick films of mCP(1-x)Bebq₂(x) doped with 5 vol.% RP411 were deposited with the values of x = 0, 0.25, 0.5, 0.75 and 1. For the sake of convenience, the fabricated devices are named as device A, device B, device C, device D, and device E according to the values of x = 0, 0.25, 0.5, 0.75 and 1, respectively. Next, the electron transport double layers of bis(10-hydroxy-benzo[h]quinolino)beryllium (Bphen) with thickness of 250 Å and ET137(electron transport material from SFC Co.) [9, 10] with thickness of 250 Å were sequentially deposited. Finally, 10 Å -thick LiF and 1200 Å -thick Al were successively deposited as a cathode.

The electroluminescent characteristics such as current density, luminance, luminous efficiency, electroluminescence(EL) spectra, and CIE color coordinates were measured with a Polaronix M6100 test system (McScience) and a CS-1000 spectro-radiometer (Konica Minolta) in a dark condition.

Results and Discussion

The energy diagram of the used materials is shown in Fig. 1. In Fig. 1, the use of DNTPD with a highest occupied molecular orbital (HOMO) level of 5.1 eV makes the hole injection easy from anode of ITO. The hole mobility and triplet energy of TAPC are 0.01 cm²/Vs and 2.87 eV, respectively [11]. The lowest unoccupied molecular orbital (LUMO) barrier of about 0.6 eV at the interface of TAPC and EML is sufficiently large to confine the electrons injected from cathode in the emitter. Therefore, the TAPC can be used as a good hole transport material in the device structure due to the high hole mobility and good electron

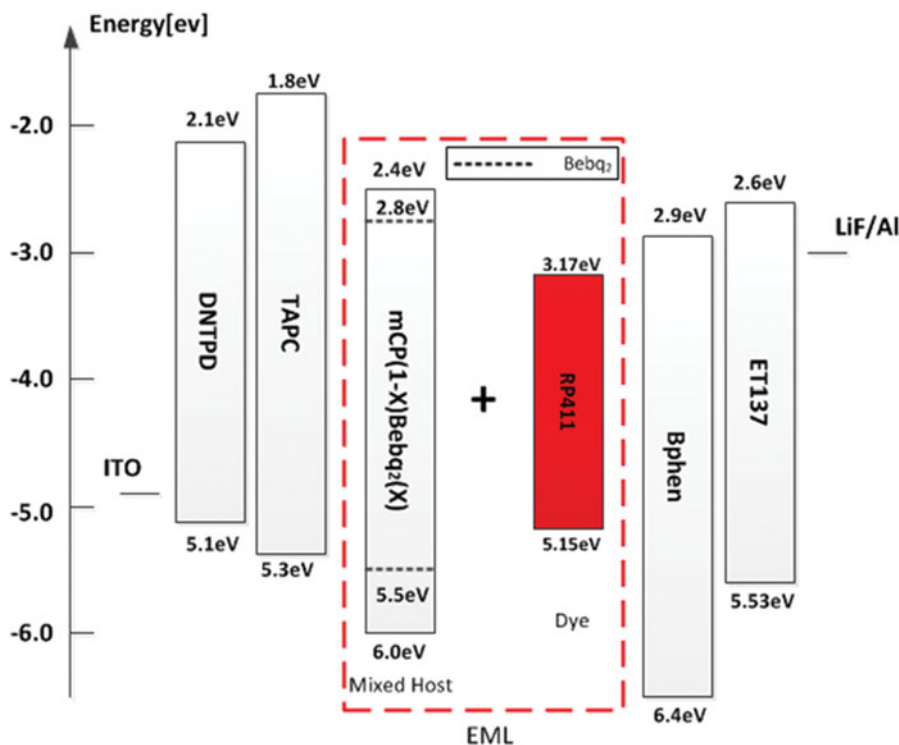


Figure 1. Energy diagram of the used materials.

confinement. Moreover, the excitons of 1.98 eV from the phosphorescent dye of RP411 can be conserved well in the hole transport layer due to the high triplet energy of TAPC.

The holes injected from anode into the emitter can be confined well by the Bphen of electron transport double layers due to a large HOMO barrier of about 0.4 eV at the interface of Bphen and EML. The triplet energy of Bphen [12], is also higher than that of RP411, so that the excitons can be also protected well in this region. The role of ET137 in the electron transport double layers helps to enhance electron injection into the emitter by ballistic transport.

The use of Beq₂ as a co-host with mCP enhances the electron injection into the emissive layer due to the lowering of LUMO barrier at the interface of Bphen and EML, and aids the electron transport in the EML due to its relative high electron mobility of about 10^{-4} cm²/Vs [13]. In addition, quantum efficiency can be greatly improved by the good energy transfer from Beq₂ to RP411 compared to that of the device with a host of mCP only.

Figure 2 shows the photoluminescence (PL) spectra of mCP/ Beq₂ and the UV-vis absorption and PL spectra of RP411. As shown in Fig. 2, the wide overlap between the PL spectra of mCP/Beq₂ and the UV-vis absorption spectra of RP411 makes it possible for the mCP/Beq₂:RP411 to be a good host-dopant system for a red emission. Chemical structures of mCP and Beq₂ are shown in the inset of Fig. 2.

Current density-voltage (J-V) and luminance-voltage (L-V) characteristics of the fabricated devices are shown in Fig. 3 and Fig. 4, respectively. The shape of luminance-voltage curves of Fig. 3 is similar to the shape of current density-voltage curves of Fig. 4 because

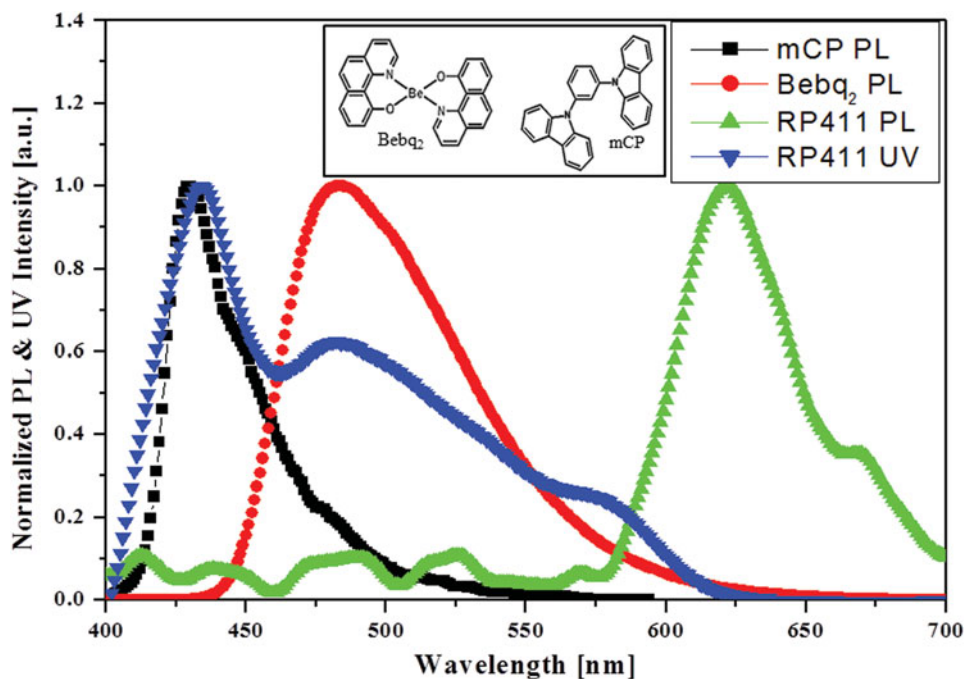


Figure 2. Photoluminescence (PL) spectra of mCP/ Bebq₂ and UV-vis absorption and PL spectra of RP411 with the inset of chemical structures of mCP and Bebq₂.

the OLEDs are driven by current injection. Current density and luminance at a given voltage showed the highest values in the device B with a host of mCP(0.75)Bebq₂(0.25). The current density and luminance of device B were 340 mA/cm² and 23,350 cd/m² under an applied voltage of 10 V, respectively. This means that the total charge transport of hole and

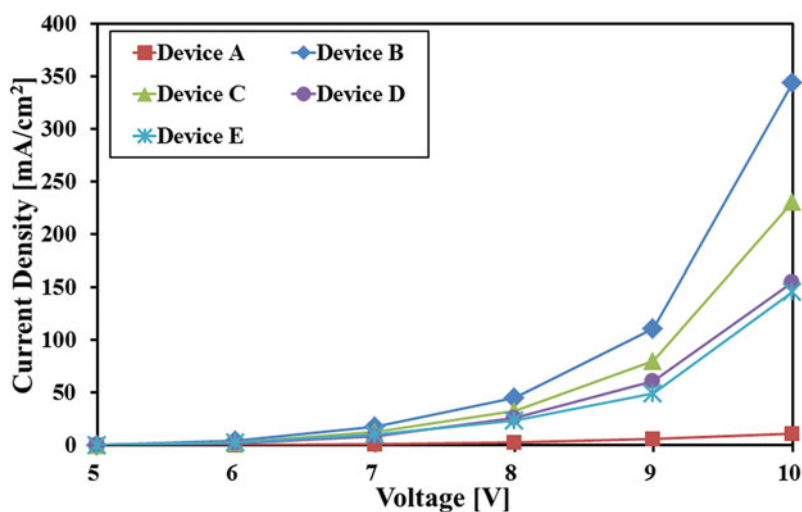


Figure 3. Current density-voltage (J-V) characteristics of the fabricated devices.

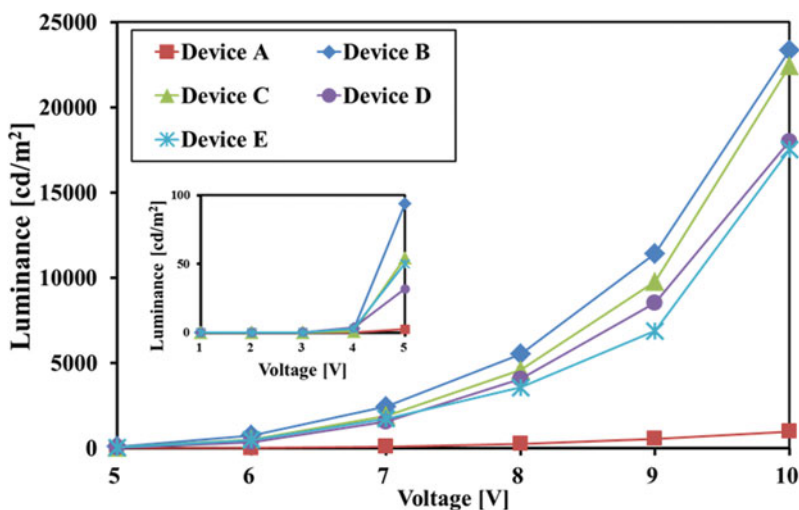


Figure 4. Luminance-voltage (L-V) characteristics of the fabricated devices.

electron becomes the largest when the ratio of Bebq_2 is about 0.25 in the mixed host of mCP and Bebq_2 .

Luminous efficiency is a very important parameter to evaluate the electroluminescent characteristics of light emitting devices. There are two kinds of luminous efficiency: current efficiency and power efficiency. The current and power efficiencies of the fabricated devices are shown in Fig. 5 and Fig. 6, respectively. We can calculate the current efficiency by an equation of L/J and the power efficiency by an equation of $\pi L/JV$ if the current density (J)-voltage (V)-luminance (L) relationship is known. In Fig. 5 and Fig. 6, the highest current and power efficiencies below 9V are shown in the device D with a host of mCP(0.25) Bebq_2 (0.75). This means that the exciton formation from an injected current and the energy

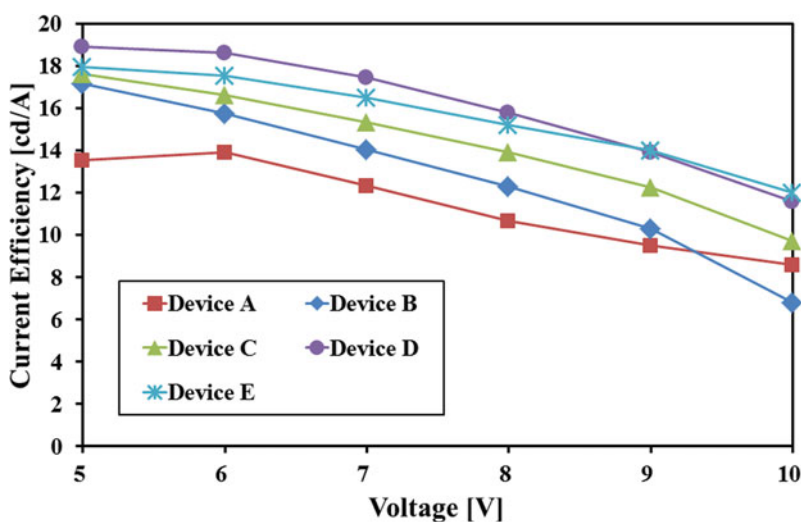


Figure 5. Current efficiency-voltage characteristics of the fabricated devices.

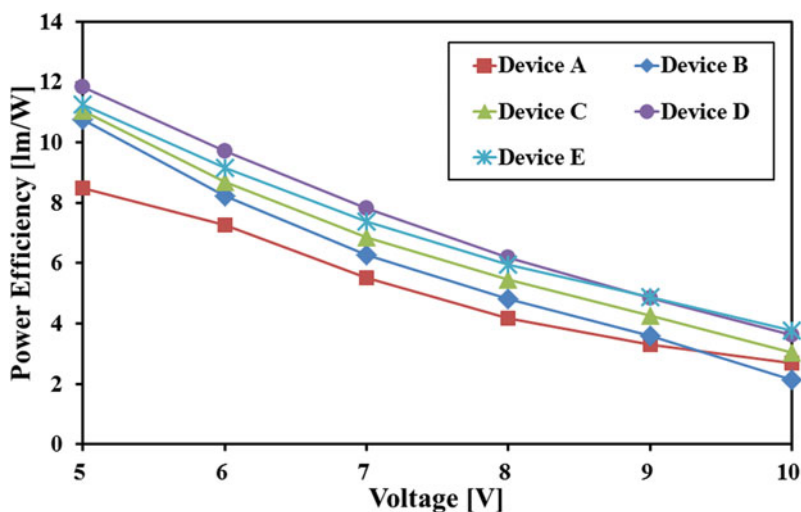


Figure 6. Power efficiency-voltage characteristics of the fabricated devices.

transfer from host to dye are optimized when the ratio of Bebq₂ is about 0.75 in the mixed host of mCP and Bebq₂. The maximum current and power efficiencies obtained in the device D were 19 cd/A and 12 lm/W. Quantum efficiency is also used to evaluate the electroluminescent characteristics of light emitting devices. Figure 7 shows the curves of quantum efficiencies of the fabricated devices according to the applied voltages. The maximum external quantum efficiency of 20% was also obtained in the device D.

In consideration of both luminance and quantum efficiency, the figure of merit for evaluating the electroluminescent characteristics can be represented as the product of them. Figure 8 shows the curves of the product of luminance and quantum efficiency normalized

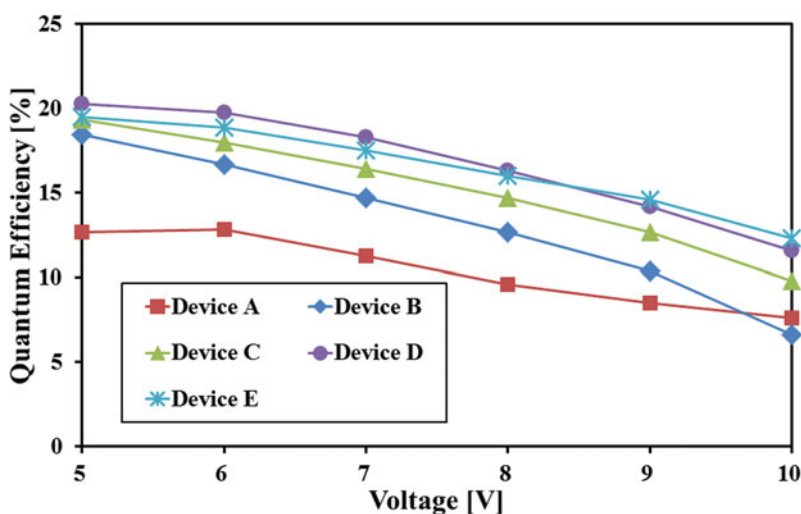


Figure 7. Quantum efficiency-voltage characteristics of the fabricated devices.

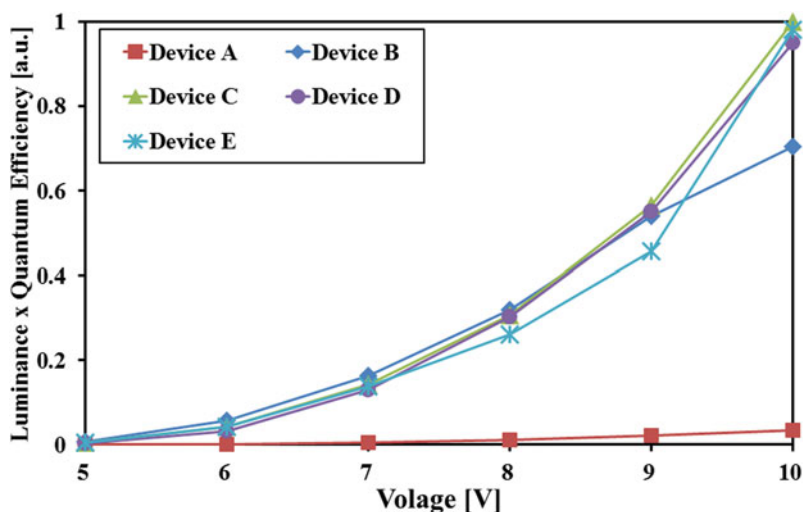


Figure 8. The product of luminance and quantum efficiency as a figure of merit according to the applied voltages.

to that of device C at 10 V. The product of luminance and quantum efficiency showed the best characteristics in the device C with a host of mCP(0.5) Beq₂(0.5) as shown in Fig. 8.

Judging from the above results, the mixture of $x = 0.5$ in the fabrication of red phosphorescent OLEDs with a mixed host of mCP(1- x) Beq₂(x) is considered to be proper to give the trade-off between luminance and quantum efficiency. The device with a host of mCP only has poor characteristics in both luminance and quantum efficiency,

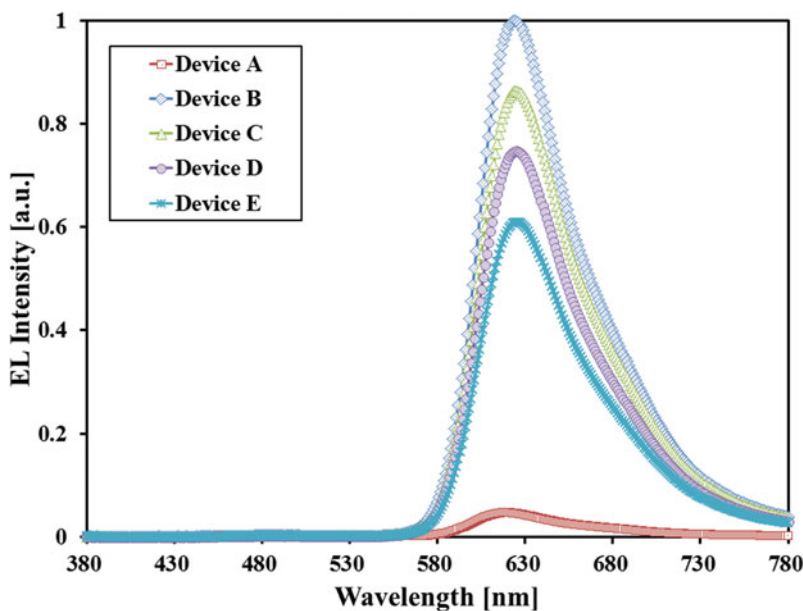


Figure 9. EL spectra of the fabricated devices under an applied voltage of 9 V.

Table 1. Key EL characteristics of the fabricated devices

Parameter	Units	Devices				
		A	B	C	D	E
Driving Voltage (at 1000 cd/m ²)	V	10	6.1	6.3	6.4	6.4
Turn-on Voltage (at 1 cd/m ²)	V	4	3.4	3.5	3.5	3.5
Luminance (at 10V)	cd/m ²	1000	23350	22420	18000	17530
Current Efficiency (at 1000 ac/m ²)	cd/A	8.6	15.5	16.1	17.9	16.9
Maximum Values						
CE	cd/A	13.9	17.1	17.6	19.0	17.9
PE	lm/A	8.4	10.7	11.0	12.0	11.2
QE	%	12.6	18.4	19.3	20.0	19.4
Peak Wavelength	nm	621	627	627	627	627
CIE	(x, y)	(0.65, 0.33)	(0.66, 0.34)	(0.66, 0.34)	(0.66, 0.34)	(0.66, 0.34)

and the device with a host of Bebq₂ only has poor luminance characteristics even though quantum efficiency is high.

EL spectra at an applied bias of 9 V are shown in Fig. 9. The peak wavelengths of EL spectra were 621 nm for device A and 627 nm for devices (B, C, D, E) similar to that of PL of RP411 in Fig. 2. This means that most of light are from triplets of RP411 with deep red emission. The color coordinates on the CIE chart were (0.65, 0.33) for the device A and (0.66, 0.34) for the devices (B, C, D, E) with color purity of more than 90%. The key EL characteristics of the fabricated devices are listed on Table 1.

Conclusions

In the fabrication of the new red phosphorescent OLEDs using a mixed host of mCP and Bebq₂ and a dye of RP411 in the emitter, the mixture ratio in a host of mCP(1-x) Bebq₂(x) on the electroluminescent characteristics were investigated according to the variation of x = 0, 0.25, 0.5, 0.75 and 1.

Among the fabricated devices, the maximum quantum efficiency was obtained in the device D with a host of mCP(0.25) Bebq₂(0.75), and the highest luminance at a given voltage was obtained in the device B with a host of mCP(0.75) Bebq₂(0.25).

In consideration of both luminance and quantum efficiency, the product of luminance and quantum efficiency as a figure of merit was the highest in the device C with a host of mCP(0.5) Bebq₂(0.5).

The device D with a host of mCP(0.25) Bebq₂(0.75) showed the maximum external quantum efficiency of 20% which resulted in the improvements of 60% in comparison with that of the device A with a single host of mCP and 3% in comparison with that of the device E with a host of Bebq₂ only. The device C with a host of mCP(0.5) Bebq₂(0.5) showed the improvements of about 30 and 1.4 times in the product of luminance and quantum

efficiency, respectively, in comparison with those of the devices (A, E) with the single hosts of mCP and BeBq₂ under the same voltage of 10 V.

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