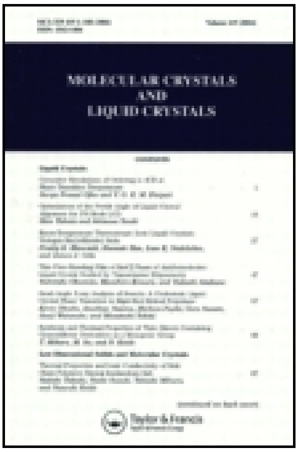

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Blue PhOLEDs Using the Mixed Host of UGH2 and Carbazole Derivative Doped with FCNIr(pic)

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New high efficiency blue light-emitting phosphorescent devices using the mixed host of p-bis(triphenylsilyl)-benzene [UGH2] and carbazole derivative like 4,4′,4″-tris(N-carbazolyl)-triphenylamine [TCTA] or N,N′-dicarbazolyl-3,5-benzene [mCP] and the dopant of 10% bis((3,5-difluoro-4-cyanophenyl)pyridine)iridium-picolinate [FCNIr(pic)] were fabricated and evaluated according to the different treatments of carbazolyl materials and the mixture ratios of TCTA/UGH2. The best result was obtained in the mixed host of UGH2 and 10% TCTA. In the devices with the mixed host of UGH2 and 10% TCTA, the addition of two exciton blocking layers of mCP and tris-[3-(3-pyridyl)mesityl]borane [3TPYMB] resulted in the improvement of current efficiency and color quality.

The device with organic structure of N,N′-diphenyl-N,N′-bis-[4-(phenyl-m-tolylamino)-phenyl]-biphenyl-4,4′-diamine [DNTPD]/N,N′-di(1-naphthyl)-N,N’-diphenylbenzidine [NPB]/mCP/(UGH2, 10% TCTA) doped with 10% FCNIr(pic)/3TPYMB/4,7-diphenyl-1,10-phenanthroline [Bphen] showed the maximum current efficiency of 9.5 cd/A and color coordinates of (0.16, 0.24) on the Commission Internationale de I’ Eclarirage (CIE) chart.

Keywords Blue phosphorescence; efficiency; mixed host; TCTA; UGH2

Introduction

Though the blue phosphorescent organic light-emitting diodes (PhOLEDs) are essential for the development of all phosphorescent active matrix OLEDs, highly efficient blue PhOLEDs are hard to obtain due to the large energy gap of dopants, which lead to insufficient carrier injection and exciton confinement. In the blue PhOLEDs, triplet of dopant should be larger than 2.7 eV for blue emission. Therefore, the use of host with wide energy gap is necessary because triplet of host should be larger than that of dopant in the host-dopant system of an emissive layer. One of the well-known host materials in the blue PhOLEDs is the UGH2, which has been used for blue PhOLEDs because of its high energy gap and triplet energy [1–3]. However, the HOMO level of 7.2 eV is too high to inject holes from a hole transport layer into an emitter. Iridium(III) bis((4,6-difluorophenyl)-pyridinate-N,C2′)picolinate [FIrpic] is the most well-known blue phosphorescent dopant [4]. Kawamura et al. demonstrated that the photoluminescent internal quantum yield of a blue emitter of FIrpic could approach nearly 100% when doped into the wide energy gap

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*Address correspondence to J. G. Jang, Department of Electronics Engineering, Dankook University, San 29, Anseo-dong, Cheonan, Chungnam 330-714, Korea (ROK). Tel.: (+82)41-550-3545; Fax: (+82)41-550-3589. E-mail: semicgk@dankook.ac.kr host of mCP [5]. However, the theoretical electrophosphorescence is difficult to carry out because of the lack of highly efficient carrier transport materials with wide energy gap for sufficient carrier injection and exciton confinement. Furthermore, the color purities of the FIrpic-based devices were poor with vertical coordinates of more than 0.3 on the CIE chart. The quantum efficiency remarkably decreases due to carrier injection loss, nonradiative relaxation of excitons, triplet-triplet annihilation at high current density, etc. [6,7]. Triplets have rather long lifetime so that they may diffuse to the neighbor layers by passing through an emissive layer. This effect also results in the declination of quantum efficiency and color purity due to energy transfer and relaxation of excitons outside an emissive layer. Therefore, the proper structure design of PhOLEDs which can confine the triplets in an emitter is extremely important to obtain high quantum efficiency.

In this work, new blue PhOLEDs using the mixed host of UGH2 and carbazole derivative doped with 10% FCNIr(pic) as a volume ratio in an emitter which have the hole injection layer of DNTPD and the hole transport layer of NPB and the electron transport layer of Bphen were fabricated and their electroluminescent characteristics according to the kinds of carbazole derivatives (TCTA, mCP) mixed into the UGH2 and the ratios of TCTA to UGH2 especially in the (UGH2, TCTA)-host devices were compared. The FCNIr(pic) has a wide triplet of 2.72 eV for deep blue emission due to a strong electron withdrawing CN substituent besides F units in the molecular scheme [8]. The effect of adding the exciton blocking layers of mCP and 3TPYMB on both sides of the emitter on the improvement of device performance was also investigated in the (UGH2, 10% TCTA)-host device. The molecular schemes of hosts and dopant used in the experiments are shown in Fig. 1.

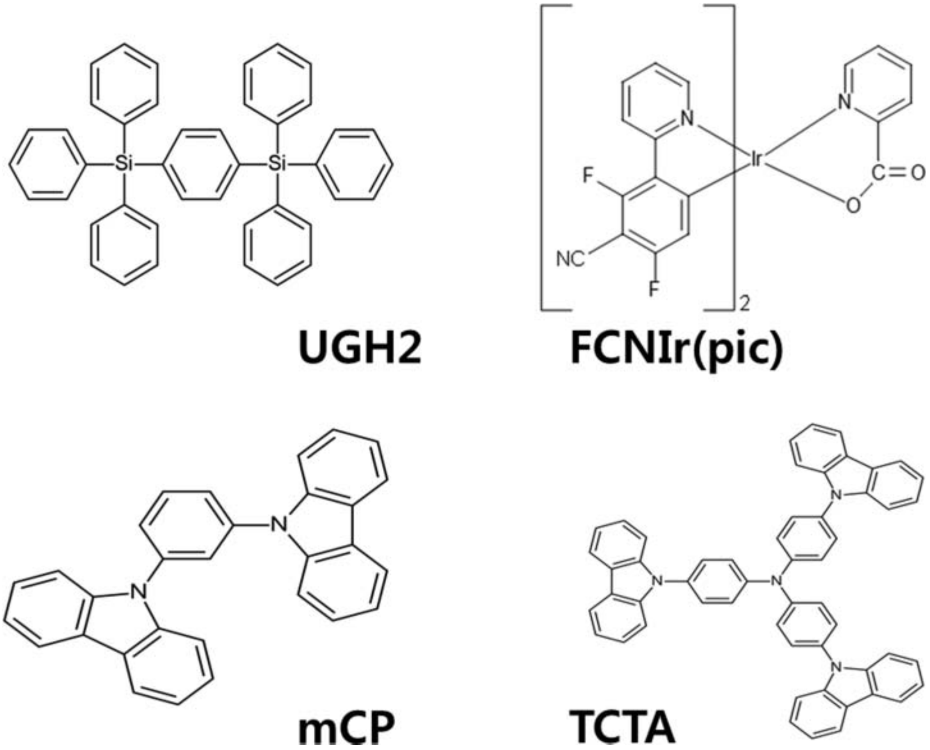

Figure 1. Molecular schemes of hosts and dopant used in the experiments.

Experimental Procedure

The substrates with an ITO (indium tin oxide) anode of 12 Ω/sq on glass were cleaned by ultrasonic cleaning process with acetone and isopropyl alcohol. The remaining solvent was removed by soft-baking for 10 minutes at 100 °C. To improve the surface morphology of ITO transparent electrode film, the substrates were plasma-treated at 150W for two minutes under 8 mTorr pressure of O_2/Ar. The plasma treatment before deposition of the first organic layer is expected to reduce the energy barrier for hole injection from anode and remove the surface contaminants.

As a sequence of process in the fabrication of the devices (1–5), the DNTPD with thickness of 400 Å were firstly deposited as a hole injection layer (HIL). And then, a 200 Å-thick film of NPB was formed as a hole transport layer (HTL). In the deposition of a 300 Å-thick emitter, five different compositions were made according to the device classifications: the UGH2:FCNIr(pic) of 10:1 in the device 1 (D1), the UGH2:mCP:FCNIr (pic) of 10:1:1 in the device 2 (D2), the UGH2:TCTA:FCNIr(pic) of 10:1:1 in the device 3 (D3), the UGH2:TCTA:FCNIr(pic) of 10:2:1 in the device 4 (D4), the UGH2:TCTA: FCNIr(pic) of 10:0.5:1 in the device 5 (D5) as a volume ratio. Next, the Bphen with thickness of 400Å was deposited as an electron transport layer (ETL). Finally, 10Å-thick LiF and 1200Å-thick Al were successively deposited as a cathode. The device 6 (D6) has the exciton blocking layers (EBLs) of a 200 Å-thick mCP and a 100 Å-thick 3TPYMB on both sides of the emitter from the device 3. The energy diagram of the used materials is shown in Fig. 2.

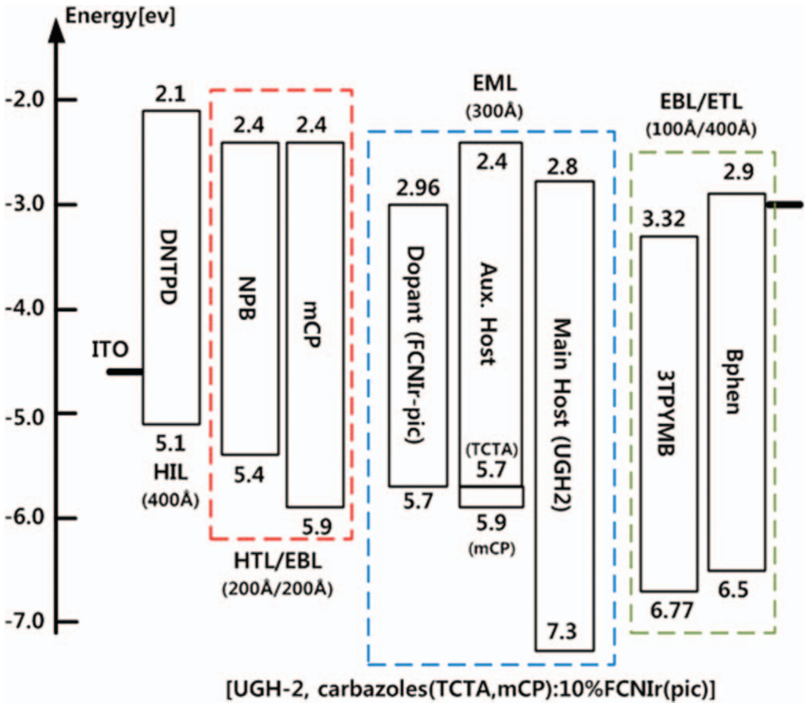

Figure 2. Energy diagram of the used materials.

Results and Discussion

The DNTPD is well known as a HIL with the HOMO level of 5.1 eV and the LUMO level of 2.1 eV in the energy diagram of Fig. 2 [9]. The electrons from the anode of indium tin oxide (ITO) are easily injected into the DNTPD because its highest occupied molecular orbital (HOMO) level is similar to Fermi level of ITO. The NPB as a HTL has the HOMO level of 5.4 eV and the LUMO level of 2.4 eV. The triplet energy of mCP is 2.9 eV [10] which is larger than that of FCNIr(pic) so that triplets can be effectively confined without diffusion loss into the HTL. Therefore, the mCP can be used as a good EBL for high efficiency blue PhOLEDs. Both Bphen and 3TPYMB have been used as a material of ETL in the conventional OLEDs. The triplet energy of Bphen is 2.6 eV [11] so that the Bphen can not sufficiently confine the excitons. On the other hand, 3TPYMB has a triplet energy of 2.87 eV which is higher than the exciton energy of FCNIr(pic) and a deep HOMO level of 6.77 eV [12]. Therefore, the use of 3TPYMB as an ETL can expect an effective confinement of the excitons as well as the injected holes in the emissive layer. However, the electron mobility of 3TPYMB is much lower than that of Bphen [13]. To solve these problems, the double layer of 3TPYMB/Bphen can be used as an EBL/ETL. The addition of a properly thick 3TPYMB to Bphen in the ETL structure can improve the electroluminescent characteristics in the blue PhOLEDs because the excitons and the injected holes are effectively confined in the emissive layer without a serious increase of current resistance.

Figure 3 shows the UV-visible absorption and photoluminescence (PL) spectra of FCNIr(pic). The PL emission peak of FCNIr(pic) was observed at 460 nm with a vibrational peak at 485 nm. The FCNIr(pic) shows a deep blue emission due to the strong electron withdrawing CN group in the phenyl unit of main ligand. The hole injection and charge balance in the emitter can be improved in the use of the mixed host of UGH2 and carbazole derivative (mCP or TCTA) compared to the use of the UGH2 only because the carbazole derivatives are hole transporting materials with a higher triplet energy of 2.9 eV [14] than

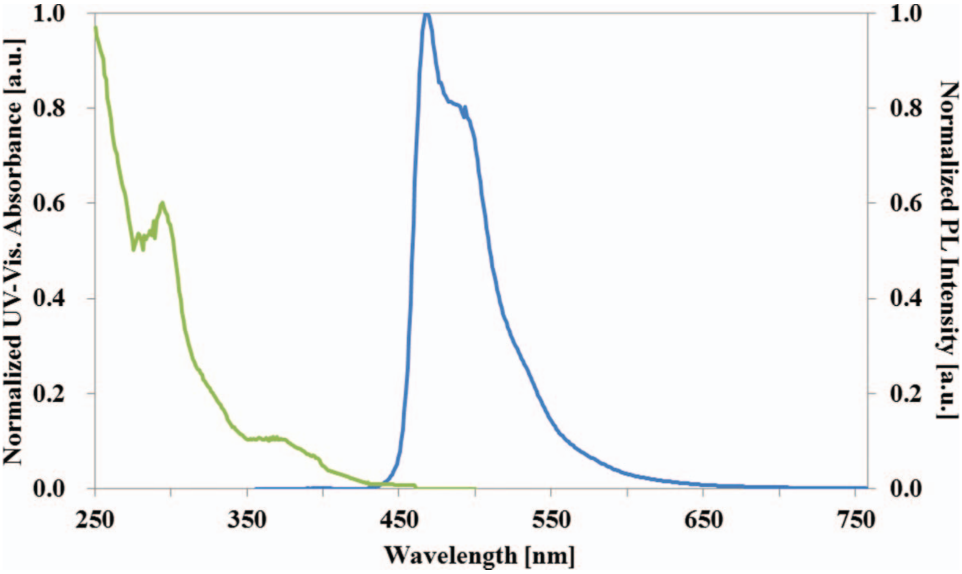

Figure 3. UV-vis absorption and photoluminescence spectra of FCNIr(pic)-dopant.

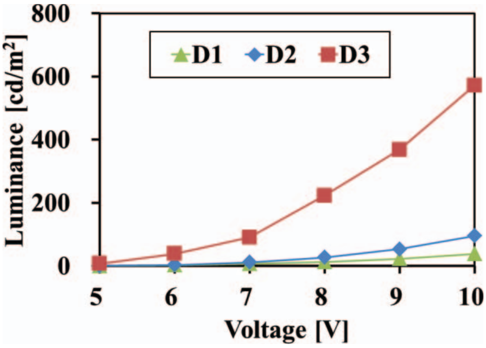

Figure 4. Luminance-voltage characteristics of the devices (1–3).

that of FCNIr(pic) and an intermediate HOMO level between the HTL of NPB and the main host of UGH2.

The electrical properties of the devices were measured using a Polaronix M6100 test system (McScience). The optical properties such as luminance, emission spectrum and CIE color coordinates were evaluated using a CS-1000 spectro-radiometer (Konica Minolta) in a dark room. The luminance-voltage and the current efficiency-voltage characteristics of the devices (1–3) are shown in Fig. 4 and Fig. 5, respectively. The luminances under an applied voltage of 10 V were 38 cd/m^2 for the device 1, 96 cd/m^2 for the device 2 and 572 cd/m^2 for the device 3 in Fig. 4. The maximum current efficiencies were 0.3 cd/A for the device 1, 1.0 cd/A for the device 2 and 5.5 cd/A for the device 3 in Fig. 5. The remarkable improvement of electroluminescent characteristics in the devices (2, 3) compared with the device 1 comes from the use of the mixed host of UGH2 and carbazole derivative instead of the single host of UGH2 in an emitter. Though the reason that electroluminescent characteristics of device 3 with the mixed host of UGH2 and TCTA are much better than those of device 2 with the mixed host of UGH2 and mCP is not clear, the experimental results show that the mixed host of UGH2 and TCTA can be a good candidate as a new host to obtain the high efficient and blue PhOLEDs. The electroluminescence spectra at 10 V for the devices (1–6) are shown in Fig. 6. The peak wavelengths of electroluminescence of

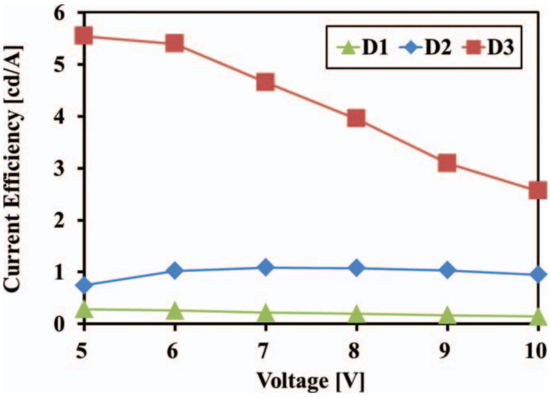

Figure 5. Current efficiency-voltage characteristics of the devices (1–3).

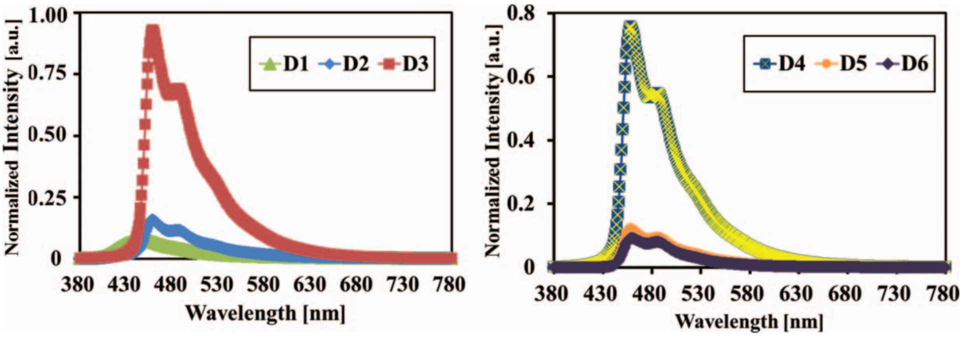

Figure 6. Electroluminescence spectra of the devices (1–6) at 7 V.

the devices (2–6) were 458 nm and 485 nm, which corresponds to the typical double peak PL emission of FCNIr(pic). The color with the first peak wavelength of 458 nm could be evaluated to be blue. On the other hand, no dopant emission could be seen but the NPB emission with a peak wavelength of around 450 nm was obtained in the device 1 with the host of UGH2 only.

As the mixed host of UGH2 and TCTA could be regarded as the best one from the above results, three kinds of devices according to the volume ratios of TCTA to UGH2 of 5%, 10% and 20% were next compared to determine the optimum ratio of TCTA to UGH2 in a mixed host of UGH2 and TCTA. Figure 7 shows the current efficiency-voltage-luminance characteristics of the devices (4, 5) compared with the device 3. The luminances under an applied voltage of 10 V were 403 cd/m^2 for the device 4 and 78 cd/m^2 for the device 5. The maximum current efficiencies were 4.7 cd/A for the device 4 and 1.4 cd/A for the device 5. The overall electroluminescent characteristics of the devices (4, 5) were inferior to those of device 3 in Fig. 7. The mixture ratio of 5% TCTA into the UGH2 in the device 5 is thought to be too low to obtain a good mixed effect of host, while the mixture ratio of 20% TCTA into the UGH2 in the device 4 is so high that electroluminescent characteristics may be declined due to exciton quenching by the molecular interaction of host materials.

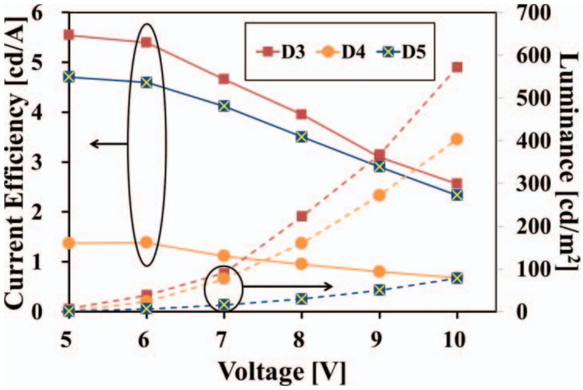

Figure 7. Current efficiency-voltage-luminance characteristics of the devices (3–5).

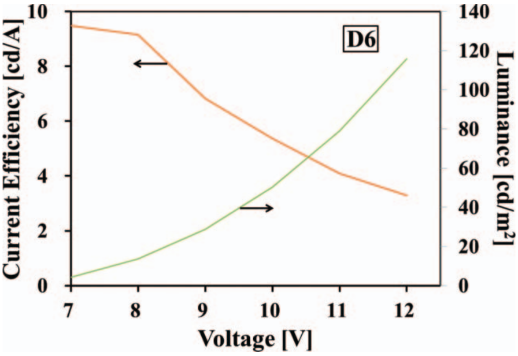

Figure 8. Current efficiency-voltage-luminance characteristics of device 6.

From the comparison of the devices (3–5), the optimum ratio of TCTA to UGH2 in a mixed host of UGH2 and TCTA could be considered to be about 10% so that the performance improvement in the (UGH2, 10% TCTA)-host device was attempted by adding the exciton blocking layers of mCP and 3TPYMB on both sides of the emitter from the structure of device 3. Figure 8 shows the current efficiency–voltage–luminance characteristics of device 6 with organic structure of DNTPD/NPB/mCP/(UGH2, 10% TCTA):10% FCNIr(pic)/3TPYMB/Bphen. In Fig. 8, the maximum current efficiency and the luminance at 10V of device 6 were 9.5 cd/A and 50 cd/m^2, respectively. The color coordinates of device 6 on the CIE chart were somewhat shifted toward a deeper blue of (0.16, 0.24) compared to (0.17, 0.25) of devices 3. Detailed device data were summarized in Table 1.

Comparing the electroluminescent characteristics of device 6 with those of device 3, the addition of two EBLs of mCP and 3TPYMB resulted in the improvement of current efficiency and color quality with a decrease of luminance. The improvement of current efficiency and color quality in the device 6 compared to the device 3 could be attributed to the effective exciton confinement in the emitter by adopting the EBLs of mCP and 3TPYMB. On the contrary, the decrease of luminance in the device 6 came from the increase of current resistance due to the addition of the EBLs. Of blue PhOLEDs with color coordinates of less than (0.16, 0.24) ever reported so far, the current efficiency of

Table 1. Electroluminescent properties of the fabricated devices

		Maxium Efficiency			
Devices	Turn-on Voltage (at 1 cd/m^2) [V]	Current Efficiency [cd/A]	Power Efficiency [lm/W]	First peak Wavelength [nm]	CIE (at 10V)
D1	5	0.3	0.2	450	(0.18, 0.15)
D2	5	1.0	0.5	458	(0.17, 0.25)
D3	5	5.5	3.5	458	(0.17, 0.25)
D4	5	4.7	3.0	458	(0.17, 0.24)
D5	5	1.4	0.9	458	(0.18, 0.26)
D6	6	9.5	4.2	458	(0.16, 0.24)

9.5 cd/A is rather high so that the structure of device 6 can be used in the development of blue PhOLEDs with high current efficiency.

Conclusions

New blue FCNIr(pic)-emitting devices using the mixed host of UGH2 and carbazole derivative like TCTA or mCP in the emitter were proposed and evaluated in this study.

In order to investigate the device performance according to the carbazole derivatives mixed into the UGH2, electroluminescent characteristics of the devices using the mixed hosts of (UGH2, TCTA) and (UGH2, mCP) were firstly compared with those of the device using the UGH2 only as a host. And then, three kinds of devices with different volume ratios of TCTA to UGH2 of 5%, 10% and 20% were fabricated to determine the optimum mixture ratio of TCTA to UGH2 in a mixed host of UGH2 and TCTA. In addition, the performance improvement in the (UGH2, 10% TCTA)-host device was attempted by adding the exciton blocking layers of mCP and 3TPYMB on both sides of the emitter.

The device with organic structure of DNTPD/NPB/mCP/(UGH2, 10% TCTA) doped with 10% FCNIr(pic)/3TPYMB/Bphen showed the maximum current efficiency of 9.5 cd/A and color coordinates of (0.16, 0.24) on the Commission Internationale de I' Eclarirage (CIE) chart.

The current efficiency of 9.5 cd/A is regarded to be sufficiently high as of a blue PhOLED.

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