

Green Phosphorescent OLED Without a Hole/Exciton Blocking Layer Using Intermixed Double Host and Selective Doping

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Abstract Simple and high efficiency green phosphorescent devices using an intermixed double host of 4, 4', 4"-tris(N-carbazolyl) triphenylamine [TCTA], 1, 3, 5-tris (N-phenylbenzimidazole-2-yl) benzene [TPBI], phosphorescent dye of tris(2-phenylpyridine)iridium(III) [Ir(ppy)₃], and selective doping in the TPBI region were fabricated, and their electro luminescent characteristics were evaluated. In the device fabrication, layers of 70 Å -TCTA/90 Å -TCTA_{0.5}TPBI_{0.5}/90 Å -TPBI doped with Ir(ppy)₃ of 8% and an undoped layer of 50 Å -TPBI were successively deposited to form an emission region, and SFC137 [proprietary electron transporting material] with three different thicknesses of 300 Å, 500 Å, and 700 Å were used as an electron transport layer. The device with 500 Å -SFC137 showed the luminance of 48,300 cd/m² at an applied voltage of 10 V, and a maximum current efficiency of 57 cd/A under a luminance of 230 cd/m². The peak wavelength in the electroluminescent spectral and color coordinates on the Commission Internationale de l'Eclairage [CIE] chart were 512 nm and (0.31, 0.62), respectively.

Key words green phosphorescent OLED, TCTA, TPBI, Ir(ppy)₃, intermixed double host, selective doping.

1. Introduction

Recently, the studies of OLEDs (organic light emitting devices) for high efficiency have been reported on a variety of structures and materials of devices.¹⁻⁵⁾ OLEDs are generally designed in a multilayer structure of inserted organics between anode and cathode which consist of hole injection layer(HIL), hole transport layer (HTL), emissive layer (EML), hole / exciton blocking layer(HBL) and electron transport layer(ETL). The basic mechanism of OLEDs consists of the formation of excitons by injection of electrons and holes from electrodes and the energy release in the emission region.

The high efficiency of OLEDs has been demonstrated by adopting phosphors such as Ir compounds in the emission region, where heavy metal effects have led to good intersystem crossing from singlets to triplets and strong phosphorescence even at room temperature.^{6,7)} However, triplet excitons have rather long lifetime so that they may diffuse to other layers by passing through the emission layer. This effect results in deterioration of luminous efficiency and color purity due to energy transfer and relaxation outside the emission layer.

Therefore, the design of phosphorescent organic light emitting devices (PhOLEDs) which can confine triplet excitons in the emission layer is extremely important to obtain high efficiency phosphorescent devices.

Various emission layers such as simple mixed, two stacked, and intermixed structure using the double host (one is a hole transporting material and the other an electron transporting material) have been reported in the PhOLEDs to solve these problems.⁸⁻¹⁰⁾ But, these devices still have some problems. In the simple mixed device, triplet excitons cannot be completely confined in the emission layer because the exciton formation zone is extended over the entire emission region. Therefore, in terms of the confinement of excitons in the emission layer, the device with a simple mixed double host requires the establishment of exciton blocking layers^{11,12)} on both sides of emission layer, but the insertion of exciton blocking layers gives rise to several disadvantages such as increase of driving voltage, complicated process, and so on. The device with two stacked double host structure has a good confinement of excitons because the exciton formation largely occurs around the interface of two hosts located in the central emission layer, but the exciton formation zone would not be wide enough to obtain high luminous efficiency due to the narrow accumulation of holes and electrons at the interface. On the other hand, it

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is possible in the intermixed double host structure to give a good confinement of excitons as well as a sufficient formation space to avoid triplet-triplet annihilation. However, if the intermixed structure involves even an additional hole and/or exciton blocking layer, the device becomes so complicated that it may not be useful in an industrial application.

In order to overcome these problems of conventional structures, a simple high efficient green PhOLED without an additional hole and/or exciton blocking layer using intermixed double host structure of TCTA/TCTA_{0.5} TPBI_{0.5}/TPBI and selective doping of Ir(ppy)₃ in the emission region was newly proposed. Ir(ppy)₃ was uniformly doped in the all region of TCTA and TCTA_{0.5} TPBI_{0.5} and selectively doped in the TPBI region. The undoped TPBI region was used to function itself as an effective blocking layer of exciton without addition of BCP [2,9-dimethyl-4,7-diphenyl-1,10-phenanthroline] or Balq [bis(2-methyl-8-quinolinolato) (p-phenylphenolato) aluminium(III)] found in conventional devices.

In this study, electroluminescent characteristics of a newly proposed device without an extra hole/exciton blocking layer were firstly compared with those of a conventional device with Balq as a HBL. Thereafter, the effect of ETL thickness variation on the electroluminescent characteristics in the newly proposed device was investigated.

2. Experimental procedure

The substrate with an indium-tin-oxide(ITO) anode of 10 Ω/sq on glass was cleaned by an ultrasonic cleaning process with acetone, methanol and isopropyl alcohol. The remaining solvent was removed by soft baking for 10 minutes at 100 °C To improve the surface morphology of the ITO transparent electrode film was plasma-treated at 150 W for two minutes with a DC bias of 37 V under 8 mTorr pressure of O₂/Ar(2/1). The plasma treatment before deposition of the first organic layer is expected to reduce the energy barrier for hole injection from anode and remove surface contaminations, and also improves the adhesion between ITO and organic layer. The organic thin layers and metallic layers were deposited under 5×10⁻⁸ Torr using in-situ method. After the substrate was moved from the plasma chamber into the high vacuum organic chamber, N,N'-diphenyl-N,N'-bis- [4-(phenyl-m-tolylamino)-phenyl] biphenyl 4,4'-damine [DNTPD] with thickness of 500 Å and N,N'-bis (1-naphthyl)-N,N'-diphenyl

-1,1'-biphenyl-4,4'-diamine [NPB] with thickness of 300 Å were successively deposited as a hole injection layer and a hole transport layer on the ITO electrode, respectively. In the formation of emission region, the layers of 70 Å-TCTA/90 Å-TCTA_{0.5} TPBI_{0.5}/90 Å-TPBI doped with Ir(ppy)₃ of 8% as a deposition rate and an undoped layer of 50 Å-TPBI were deposited in order. Then, SFC137 with different thicknesses of 300 Å, 500 Å and 700 Å was used as an electron transport layer according to the experimental devices. Finally, 10 Å-LiF and 1200 Å-Al were successively deposited as a cathode. In the conventional device, TPBI was all doped with Ir(ppy)₃ of 8% and the thickness of SFC137 was 500 Å. Besides, Balq with thickness of 50 Å was added as an additional exciton blocking layer between TPBI and SFC137. Energy band diagram of the multilayered organic structure was shown in Fig. 1.

3. Results and discussion

Current density(J) - luminance(L) - voltage(V) characteristics of the devices were measured by a Polaronix M6100 test system (McScience). Optical properties such as emission spectrum and CIE color coordinates were investigated using a CS-1000 spectro-radiometer (Konica Minolta) in a dark room.

Fig. 2 shows the current efficiency versus luminance characteristics of the newly proposed device with 500 Å-SFC137 in comparison with those of the conventional device. Current efficiencies of the newly proposed device were not inferior to those of the conventional device in a wide range of luminescence as shown in Fig. 2. The maximum current efficiencies of the newly proposed device and the conventional device were 57 cd/A and 51 cd/A, respectively. Simpler structure without any deterioration of performance makes the newly proposed

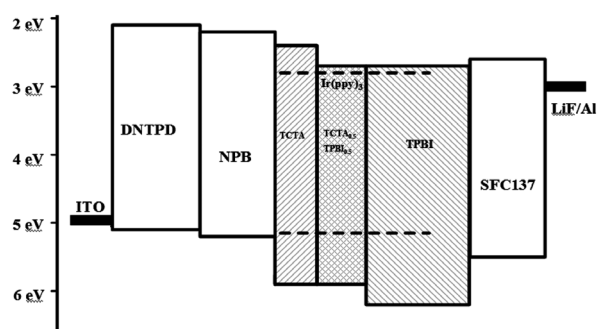


Fig. 1. Energy band diagram of the multilayered organic structure.

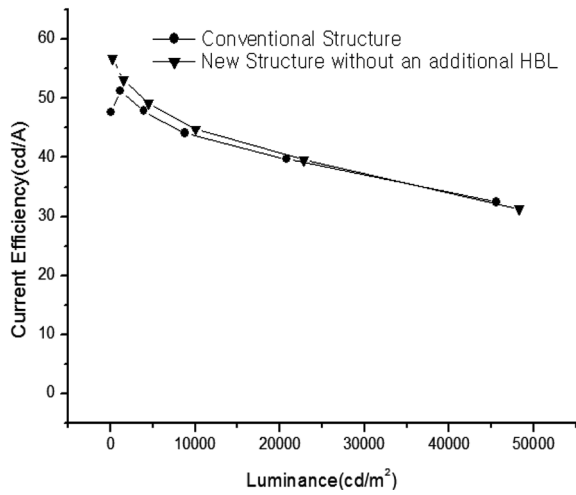


Fig. 2. Current efficiency versus luminance characteristics for the newly proposed device with SFC137 of 500 Å and the conventional device.

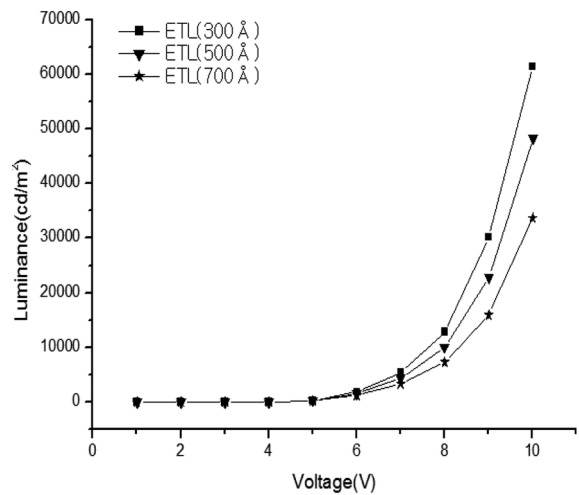


Fig. 4. Luminance versus voltage characteristics according to the thickness variation of ETL.

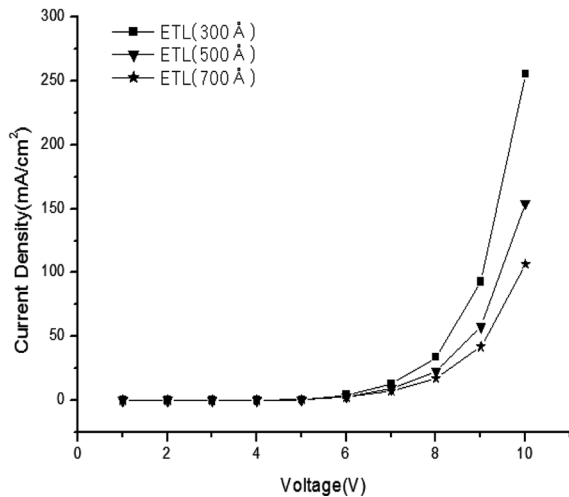


Fig. 3. Current density versus voltage curves according to the thickness variation of ETL.

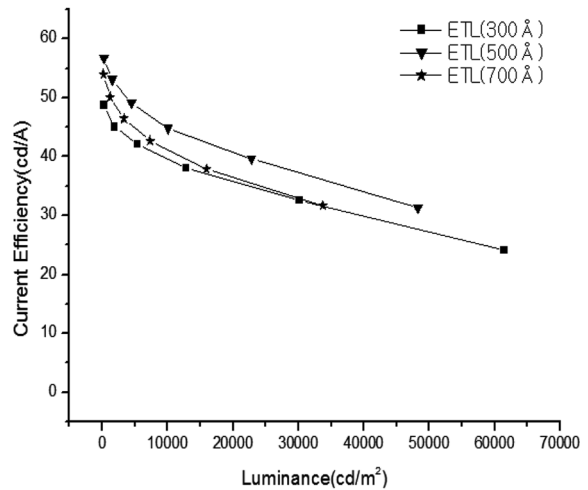


Fig. 5. Current efficiency versus luminance characteristics according to the thickness variation of ETL.

device very attractive in the device application. It is thought that good performance of this device even without any additional HBL could be obtained by the adoption of intermixed double host structure from TCTA and TPBI and selective doping in the TPBI region. The newly proposed device does not need any additional HBL because the undoped layer of TPBI has a function of effective hole/exciton blocking layer itself.

The current density vs voltage characteristics and the luminance vs voltage characteristics for the newly proposed devices with different thicknesses of ETL were shown in Fig. 3 and Fig. 4, respectively. As shown in Fig. 3 and Fig. 4, both current density and luminance were the highest in

the device with 300 Å-SFC137 as an ETL at a given voltage. On the other hand, the lowest values of current density and luminance were obtained in the device with 700 Å-SFC137. This fact explains that the thicker ETL leads to the higher resistance resulting in the lower current density and less luminance. The devices deposited with SFC137 of 300 Å, 500 Å and 700 Å showed current densities of 260 mA/cm², 150 mA/cm², 110 mA/cm² and luminances of 61,500 cd/m², 48,300 cd/m², 33,700 cd/m² at an applied voltage of 10 V, respectively.

Current efficiency is an important parameter to evaluate the electroluminescent characteristics of a light emitting device. The current efficiency can be calculated by the relation of L/J if the current density-voltage and luminance

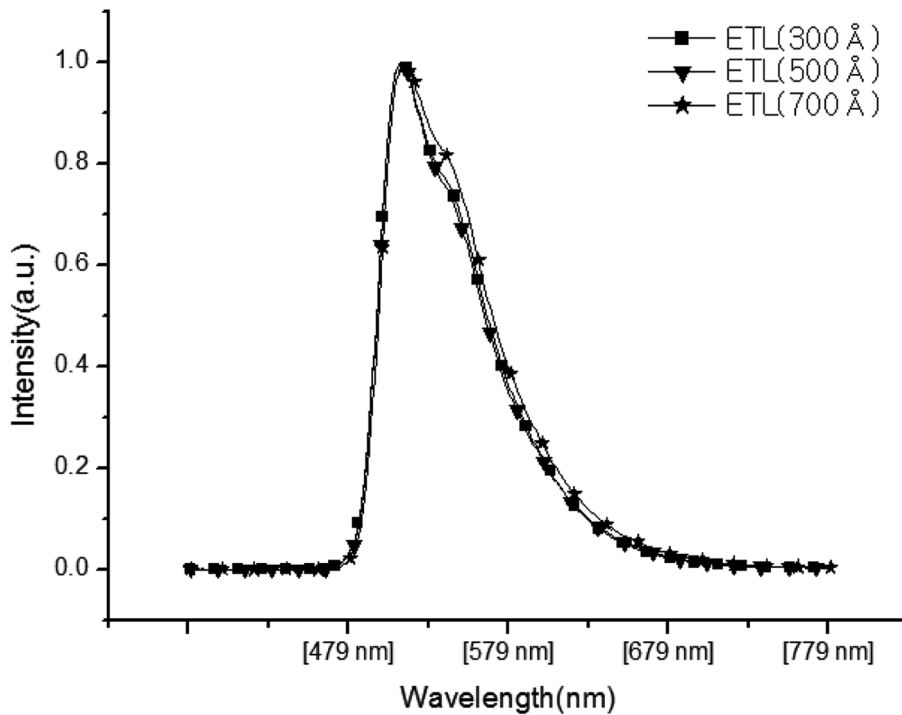


Fig. 6. Electroluminescent spectral.

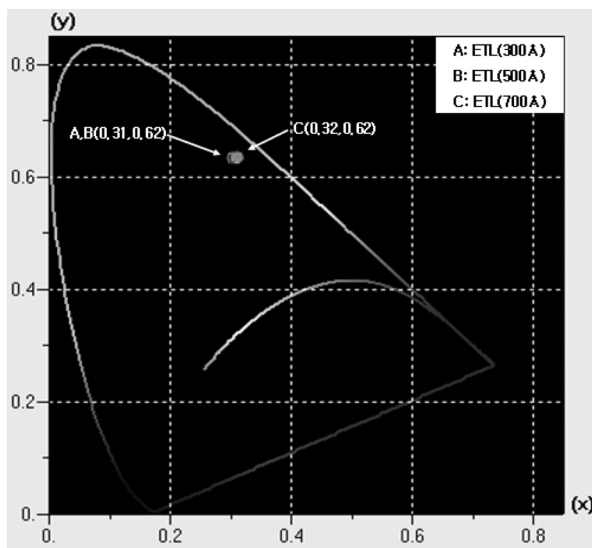


Fig. 7. CIE color coordinates.

-voltage relationships are known, where L (cd/m^2) and J (A/m^2) are luminance intensity and current density, respectively. Fig. 5 shows the current efficiency vs luminance characteristics of the devices with different thicknesses of SFC137. Among the experimental devices, the best current efficiency was obtained in the device with SFC137 of 500 Å. The device with SFC137 of 500 Å showed the maximum current efficiency of 57 cd/A under a luminance of 230 cd/m^2 .

The current efficiencies of the devices with SFC137 of 300 Å, 500 Å and 700 Å were 40 cd/A , 45 cd/A , 41 cd/A under a luminance of 10,000 cd/m^2 , respectively. The current efficiency tended to slightly decrease according to the increase of luminance for each device.

The electroluminescent spectral distributions and CIE color coordinates for the fabricated devices around 7 V were shown in Fig. 6 and Fig. 7. Some important electroluminescent characteristics for the fabricated devices were summarized in Table 1. For the devices with SFC137 of 300 Å, 500 Å and 700 Å, central wavelengths of emission spectra were 512 nm, 512 nm and 524 nm in Fig. 6 and color coordinates were (0.31,0.62), (0.31,0.62) and (0.32,0.62) in Fig. 7, respectively.

4. Conclusion

Simple and high efficiency green phosphorescent devices using the emission structure of [70 Å-TCTA/90 Å-TCTA_{0.5}TPBI_{0.5}/90 Å-TPBI]:8%-Ir(ppy)₃/50 Å-TPBI(undoped) with different thicknesses of SFC137 were newly proposed and evaluated. The structure of experimental devices can be said to be relatively simple because any additional hole/exciton blocking layer is not used. And also, the intermixed structure of emission region gives a good

Table 1. Electroluminescent characteristics of the fabricated devices.

Thickness of SFC 137	Luminance at 10V	Current Efficiency (at 10,000 cd/m ²)	Max. Current Efficiency	Peak Wavelength	CIE (x,y)
300 Å	61,500 cd/m ²	40 cd/A	49 cd/A (at 260 cd/m ²)	512 nm	(0.31, 0.62)
500 Å	48,300 cd/m ²	45 cd/A	57 cd/A (at 230 cd/m ²)	512 nm	(0.31, 0.62)
700 Å	33,700 cd/m ²	41 cd/A	54 cd/A (at 200 cd/m ²)	514 nm	(0.32, 0.62)

confinement of excitons within emission region because exciton formation largely occurs in the TCTA_{0.5}TPBI_{0.5} layer. Moreover, the intermixed structure provides sufficient exciton formation space to avoid the narrow accumulation of holes and electrons around the TCTA-TPBI interface and the resultant triplet-triplet annihilation found in a stacked double host structure. Therefore, the newly proposed device in this paper will be a good candidate as one of high performance green PhOLEDs.

In the experiments, the best luminance characteristics was obtained in the device with SFC137 of 500 Å, which had the luminance of 48,300 cd/m² at an applied voltage of 10 V and the maximum current efficiency of 57 cd/A under a luminance of 230 cd/m². The good electroluminescent characteristics of this device could be obtained owing to the structural improvement which can give the effective confinement and sufficient formation space for excitons by adopting the intermixed double emission structure and selective doping in the electron transporting host and the proper treatment for electron transport layer.

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