Highly Efficient Green Phosphorescent Organic Light Emitting Diodes

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Abstract

We have developed green phosphorescent organic light-emitting diodes (OLEDs) with high quantum efficiency. Wide-energy-gap material, 1,1-bis[(di-4-tolylamino) phenyl]cyclohexane (TAPC), with high triplet energy level was used as a hole transporting layer. Electrophosphorescent devices fabricated using TAPC as a hole-transporting layer and N,N'-dicarbazolyl-4,4'-biphenyl (CBP) doped with fac-tris(2-phenylpyridine) iridium [Ir(ppy)3] as the emitting layer showed the maximum external quantum efficiency (η_{ext}) of 19.8 %, which is much higher than the devices adopting 4,4'-bis[N-(1-naphthyl)-N-phenyl-amino]biphenyl (NPB) (ηB_{ext}=14.6 %) as a hole transporting layer.

1. Introduction

Phosphorescent organic lighting emitting diodes (PhOLEDs) have received considerable attention due to their ability of highly efficient emission compared with fluorescent OLEDs.[1-4] Through harvesting both singlet and triplet excitons, the external quantum efficiency (η_{ext}) of PhOLEDs has reached above 20 % by using the optimized material systems.[5-7] For high efficiency of PhOLED, devices have to optimize considering singlet energy transfer as well as triplet energy transfer. In addition to the triplet exciton confinement by host molecules, triplet exciton diffusion into a hole-transport layer (HTL) and an electron-transport layer (ETL) should be considered, since the triplet excitons by charge carrier recombination are usually formed near such interfaces.[8] The loss of energy into the non-radiative decay levels of the HTL and ETL layers can decrease OLED efficiency. Furthermore, utilization of phosphorescent dyes as the light emitting material in OLEDs requires efficient hole, electron and exciton blocking layers. An efficient electron-blocking material must have the lowest unoccupied molecular orbital (LUMO) level lower than that of the dopant or

host material, and its' triplet energy must be high enough to prevent migration of triplet excitons from the luminescent layer.[9] Steady-state emission spectra analysis indicates that the reduction of the efficiency is also resulted from the exciton formation zone shift toward the HTL as the current density increases.[10] Thus, the higher triplet energy level is desired for HTL to block the diffusion of the triplet exciton into the HTL.

Here, we investigate significant improvement of device efficiency by using TAPC as a HTL, resulted from efficient exciton and electron blocking from emitting layer (EML) to HTL layer. Ir(ppy)$_3$ based OLEDs with TAPC HTL resulted in the maximum external quantum efficiency (η_{ext}) of 19.8 %, which corresponds to internal quantum efficiency of ~100 % by assuming that 20 % of the emitted light is extracted and the power efficiency (η_p) of 45.1 lm/W.

2. Experimental

OLEDs were fabricated by thermal evaporation onto cleaned glass substrates pre-coated with indium tin oxide (ITO) without breaking the vacuum. Prior to organic layer deposition, ITO substrates were exposed to UV-ozone flux for 10 min following degreasing in aceton and isopropanol. All organic layers were grown by thermal evaporation at the base pressure of $<5\times10^{-8}$ Torr in the following order: HTL/EML/hole blocking layer (HBL)/ETL/cathode. 40-nm-thick 4,4'-bis[N-(1-naphthyl)-N-phenyl-amino]biphenyl (NPB) or TAPC was used as the HTL, 30-nm-thick *N,N'*-dicarbazolyl-4,4'-biphenyl (CBP) doped with 6 wt.% *fac*-tris(2-phenylpyridine) iridium [Ir(ppy)$_3$] as the EML, 10-nm thick 2,9-dimethyl-4,7-diphenyl-1,10 phenanthroline (BCP) as the HBL and 40-nm-thick tris-(8-hydroxyquinoline) aluminum (Alq$_3$) as the ETL, respectively as shown in Figure 1. Finally, the cathode

consisting of a 1-nm-thick LiF and a 100-nm-thick layer of Al were deposited onto the sample surface. Figure 1 shows the structure of the devices and the materials used in this study. Current density-voltage-luminescence (J-V-L) characteristics of the OLEDs were measured simultaneously using a Keithley 2400 programmable source meter and SpectraScan PR650 (Photo Research).
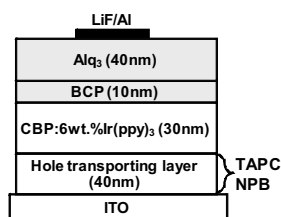
Figure 1. Structure of electrophosphorescent device.

3. Results and discussion

Quantum efficiency and power efficiency of the devices are displayed in Figure 2 (a) and summarized in Table 1. When we employ TAPC as a HTL, the η_{ext} reaches to 19.8 % at 430 cd/m^2 and 0.63 mA/cm^2. The control devices with a NPB HTL showed the maximum η_{ext} of 14.6 % and η_p of 41.7 lmW (at 1.7 cd/m^2 and 0.004 mA/cm^2), which is similar to the recently reported results.[3-5, 10]

Table 1. Summary of device performances.

HTL	η_{ext} [%]	η_p [lm/W]	η_{ext}[a] [%]	η_p[a] [lm/W]	Volt.[b] [V]	Volt.[a] [V]
TAPC	19.8	45.1	19.1	27.4	4.0	7.7
NPB	14.6	41.7	9.8	14.8	4.3	7.3

[a] measured at 1000 cd/m^2. [b] measured at 10 cd/m^2.

Normalized electroluminance (EL) spectra of the devices at different current densities are shown in Figure 2(b, c) in a logarithmic scale. It is noteworthy that the emission around 440 nm grows with increasing current density in control device with NPB HTL, and the emission begins to appear at the current density of 0.1 mA/cm^2, where the efficiency begins to be reduced. Since the emission is consistent with the emission from NPB, the *roll-off* in the quantum efficiency with increasing current density for the control device seems to be related to the exciton

formation in the NPB, the HTL.[10] However, the device with TAPC HTL shows no emission of TAPC layer up to 10 mA/cm^2, resulting in the higher quantum efficiency at high current density.
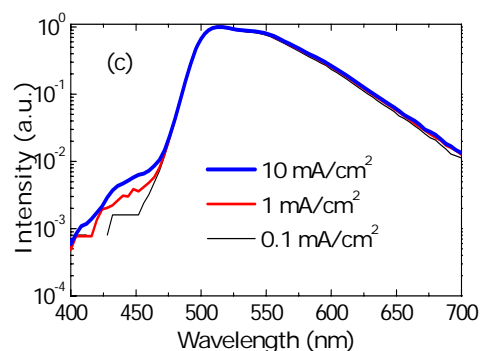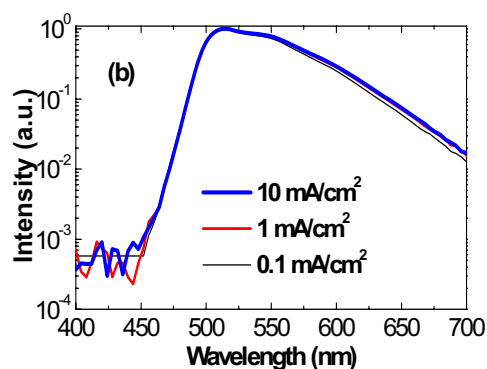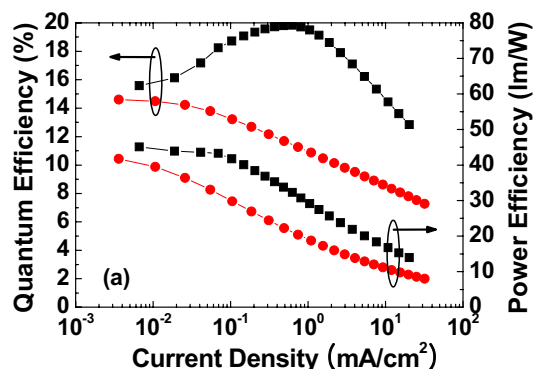
Figure 2. The external quantum efficiency and power efficiency of PhOLEDs with a TAPC (black symbols) and NPB (red symbols) HTL (**a**). Normalized electroluminescent spectra of PhOLEDs with a TAPC (**b**) and NPB (**c**).

The reason why the high quantum efficiency could be obtained from the devices with TAPC HTL can be

understood based on the energy level alignment of the layers in the devices. The LUMO levels were estimated by assuming that the HOMO-LUMO gap is equal to the optical energy gap as determined from absorption spectra.[11] Here we note the following reasons to contribute to the high quantum efficiency: The lower LUMO level of the TAPC (2.0 eV) than that of NPB (2.3 eV) leads to better confinement of electron within the EML; the triplet energy level of the TAPC (2.87 eV) is higher than that of NPB (2.29 eV) and Ir(ppy)$_3$ (2.42 eV), facilitating efficient exciton blocking from the EML to HTL. Therefore, the increased confinement of electron within the EML and efficient blocking of exciton from the EML to HTL explain the increased quantum efficiency of the device which adopted the TAPC as a HTL.
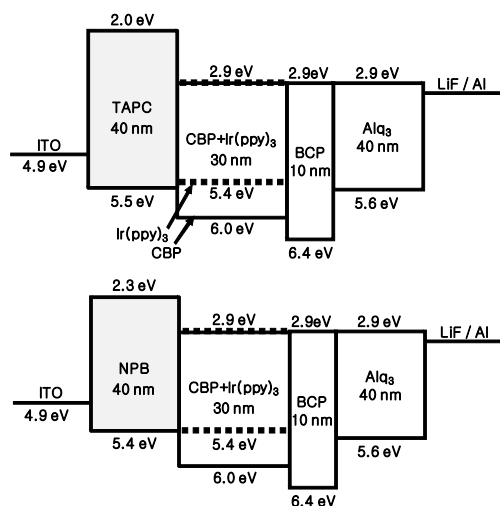

Figure 4. Energy level diagram of the device.

4. Summary

We have demonstrated significant improvement of device efficiency by using the TAPC as a HTL, resulted from efficient exciton and electron blocking from the EML to TAPC layer. The maximum η_{ext} of 19.8 % was achieved by adopting the TAPC HTL based PhOLEDs, which are much higher than the devices adopting NPB (maximum η_{ext}=14.6 %). Furthermore, the η_{ext} of 19.1 % was acheived at the luminescent intensity of 1000 cd/m^2, compared to the device with NPB HTL (η_{ext}=9.8 %). This is a promising approach for display and solid-state lighting application.

Acknowledgement

5. References

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