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High efficiency red phosphorescent organic light-emitting diodes with single layer structure

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ABSTRACT

We report simple red phosphorescent devices comprising single layer configuration using a tris[1-phenylisoqunolinato-C2,N]iridium (III) ($Ir(piq)_3$) dopant concentration as low as 1 wt.% in mixed host systems. The driving voltage of 5.4 V to reach the brightness of 1000 cd/m² in the mixed emitting layer of *N*,*N*'-diphenyl-*N*,*N*'-bis(1,1'-biphenyl)-4,4'-dia-mine (NPB) and bis(10-hydroxybenzo[h]quinolinato)beryllium (Bebq₂) is reported. Maximum current and power efficiency values of 9.44 cd/A and 10.62 lm/W are obtained in this single layer structure PHOLEDs, respectively. It is believed that the direct injection of charge carriers to the organic layer and high carrier mobility therein are the keys to the simplification of the device structure.

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1. Introduction

Organic light-emitting devices (OLEDs) have made significant stride in recent days [1] and the technology has already been commercialized to mobile flat panel display applications. In phosphorescent OLEDs (PHOLEDs), required for high brightness and display applications, almost maximum theoretical limit of external quantum efficiency (EQE) has been achieved in several color devices [2–4]. Thermal evaporation technique and complicated fabrication process consisting of multiple layers for charge carriers balancing and exciton confinement [5–7] are employed in highly efficient phosphorescent OLEDs. In order to overcome such complex device architecture, many good approaches are enduring until now. High efficiency devices with pure organic bi-layered OLEDs have been reported by several researchers [8–10]. Furthermore, bi-layered devices consisting of an organic single layer with a buffer layer on the electrode have also been reported without any significant improvement of the device performances [11–14]. However, truly organic single layered approach is almost rare. To date, only an exclusive article on the red emitting PHOLED single layer device with a tris[1-phenylisoqunolinato-C2,N]iridium (III) (Ir(piq)₃) (21 wt.%) doped in TPBi (100 nm) with low values of current and power efficiencies under 3.7 cd/A and 3.2 lm/W at 1 cd/m² have been reported, respectively [15].

In this paper, we have demonstrated efficient and simple red PHOLEDs with only single organic layer using thermal evaporation technique. The key to the simplification is the direct injection of holes and electrons into the mixed host materials through electrodes. In conventional OLEDs, usually the Fermi energy gap between cathode (\sim 2.9 eV) and surface treated anode (\sim 5.1 eV) is about 2.0–2.2 eV which is close to the red light emission energy (1.9–2.0 eV). As a consequence, red devices do not at all require any charge injection and transporter layer if the host material has proper HOMO (highest occupied molecular orbital) and



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LUMO (lowest unoccupied molecular orbital) energy levels. However, such host materials are very rare. The most suitable option to address such issues is to employ the mixed host system to adequately match the energy levels between emitting host and electrodes. Mixed host system of electron transporting and hole transporting materials to inject electrons and holes from electrodes into the organic layer without any barrier has been studied, respectively and employed for the charge balance. Thus, hole type host materials are required to have HOMO energy levels at 5.1–5.4 eV to match with the Fermi energy of surface treated ITO (5.1 eV). While 2.8-3.0 eV LUMO energy levels of electron transporting host materials are necessary to match the Fermi level of cathode. 4,4',4"-Tris(N-3-methylphenyl-N-phenyl-amino)triphenylamine(m-MTDATA) and N,N'-diphenyl-N, N'-bis(1,1'-biphenyl)-4,4'-diamine (α -NPB) were used as the hole transporting host materials. Bis(10hydroxybenzo[h]quinolinato)beryllium $(Bebq_2)$ with 2.8 eV LUMO energy was used as the electron transport host material and Ir(piq)3 was employed as a red phosphorescent guest.

2. Experimental

m-MTDATA and α -NPB as hole transporting host materials, Bebq₂ as an electron transporting host material, and $Ir(pig)_3$ as a red dopant were purchased from Gracel Corporation. To fabricate OLED devices, clean glass substrates precoated with a 150-nm-thick indium tin oxide (ITO) layer with a sheet resistance of $\sim 20 \Omega/sq$ were used. Line patterns of ITO were formed on glass by photolithography process. The ITO glass was cleaned by sonification in an isopropylalcohol and acetone, rinsed in deionized water, and finally irradiation in a UV-ozone chamber. All organic materials were deposited by the vacuum evaporation technique under a pressure of $\sim 2.0 \times 10^{-7}$ Torr. The deposition rate of organic layers was about 1 A/s. Subsequently, LiF and Al were deposited in another vacuum deposition system without breaking vacuum. Deposition rates of LiF and Al were 0.1 and 5-10 A/s, respectively. The current density-voltage (I-V) and luminance-voltage (L-V) data of red PHOLEDs were measured by Keithley 2635 A and Minolta CS-1000A, respectively. The red PHOLED emitting area was 2 mm² for all the samples studied in the present work.

3. Results and discussion

Fig. 1 shows the energy band-diagram of the single layer red PHOLEDs used in the present work. For the evaluation of single layer with different mixed host systems, the following devices were fabricated:

Device A: ITO/m-MTDATA:Bebq₂:Ir(piq)₃ [1–4 wt.%, 100 nm]/LiF (0.5 nm)/Al (100 nm). Device B: ITO/α-NPB:Bebq₂:Ir(piq)₃ [1–4 wt.%, 100 nm]/LiF (0.5 nm)/Al (100 nm).

The ratio of the hole and electron transporting hosts was fixed to 1:1. The doping concentrations were varied



Fig. 1. Energy band-diagram of the single layer red PHOLEDs.

from 1% to 4% to optimize the device performance. Table 1 shows the performance of red PHOLEDs devices comprising a single emitting layer. The current and power efficiencies values of 7.44 cd/A, and 3.43 lm/W at 1000 cd/m^2 brightness value are reported in 4 wt.% doped device A, respectively. The driving voltage (to reach 1000 cd/m^2) is 6.9 V. Very similar device performances are obtained in 2 wt.% doped device A. The optimum doping condition for device A seems to be 4 wt.% as the highest efficiency is observed at an acceptable brightness value (1000 cd/ m²). Whereas, the driving voltage, current and power efficiencies values of 5.4 V, 9.02 cd/A, and 5.25 lm/W at brightness value of 1000 cd/m² are reported in device B with 1 wt.% of optimum doping condition, respectively. Maximum current efficiency for devices A and B were appeared for the 4 and 1 wt.% of Ir(piq)₃ doped mixed hosts, respectively. The color coordinates are (0.66, 0.33) or (0.67, 0.32) for all devices. Even in 1% doped device, a good red emission color is observed. The results of device B (1 wt.%) is significantly superior to $Ir(piq)_3$ doped multilayer red PHOLEDs [16]. In our previous study, the doping concentration in PHOLEDs can be reduced until 1-2% range with higher efficiency when HOMO-HOMO and LUMO-LUMO differences between host and dopant molecules are provided within ~0.3 eV [17]. Device B displays exactly similar behavior although HOMO-HOMO gap is relatively higher than device A. However, unlike device B, similar device properties in device A regardless of doping condition from 1% to 4% are obtained. The self quenching by dopants seems to be not so serious in this device A. This indicates that the emission zone of device A is very broad and the charge balance is also relatively poor. As a result, the efficiency of device A is low compared to device B and 4% doped condition in device A seems to have a little better charge balance.

The *J*–*V*–*L* curve and efficiency characteristics of devices A and B are shown in Fig. 2. The best efficiency yields of 9.44 cd/A (EQE 14.6%) and 10.62 lm/W are noticed in the device B as shown in Fig. 2b. As seen from the results of Fig. 2a, the driving voltage in device A with m-MTDATA:Bebq_2:Ir(piq)_3 [4 wt.%] is 6.9 V at the bright-

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