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Low voltage red phosphorescent organic light-emitting devices with triphenylphosphine oxide and 4,4′-bis(2,2′-diphenylvinyl)-1,1′-biphenyl electron transport layers

Tae-Yong Kim, Dae-Gyu Moon *

Department of Materials Engineering, Soonchunhyang University, 646, Eupnae-ri, Shinchang-myeon, Asan-si, Chungcheongnam-do 336-745, Republic of Korea

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ABSTRACT

We have developed red phosphorescent organic light-emitting devices operating at low voltages by using triphenylphosphine oxide (Ph_3PO) and 4,4′-bis(2,2′-diphenylvinyl)-1,1′-biphenyl (DPVBi) electron transport layers. 4,4′-bis(*N*-carbazolyl)-1,1′-biphenyl (CBP) and tris-(1-phenylisoquinolinato-C^2,*N*) iridium(III) [$Ir(piq)_3$] were used as host and guest materials, respectively. Small voltage drops across the electron transport layers and direct injection of holes from 4,4′,4″-tris[*N*-(2-naphthyl)-*N*-phenyl-amino]-triphenylamine (2-TNATA) hole transport layer into the $Ir(piq)_3$ guests are responsible for the high current density at low voltage, resulting in a high luminance of 1000 cd/m^2 at low voltages of 2.8–3.0 V in devices with a structure of ITO/2-TNATA/CBP:$Ir(piq)_3$/DPVBi/Ph_3PO/LiF/Al.

1. Introduction

Phosphorescent organic light-emitting devices (PHOLEDs) have been interested in display and lighting applications because they can provide a high internal quantum efficiency by harvesting both electro-generated singlet and triplet excitons [1,2]. Although the green-emitting organic electrophosphorescent devices with nearly 100% internal quantum efficiency have been demonstrated [3], high driving voltage is still one of drawbacks in PHOLED applications. The PHOLEDs typically have multilayer structures composed of large band gap materials, thereby limiting the injection of carriers at organic/organic and organic/electrode interfaces. In addition, the transport of injected carriers is limited by the low mobility of carrier transport layer. Furthermore, the electron mobility of electron transport layer is generally much lower than the hole mobility of hole transport one. Kido et al. [4] proposed the electrical doping of low work function metals such as Li and Sr into the electron transport layer. These dopants create free electrons in the electron transport layer, resulting in low injection barrier and high electron conductivity. Pfeiffer et al. [5] demonstrated the low voltage green PHOLEDs using a *pin* structure [6] where both electron and hole transport layers are doped with organic or inorganic dopants. On the other hands, Park et al. [7] reported the red PHOLEDs with an operating voltage of 4.5 V at 1000 cd/m^2, without using an electrically doped transport layer. They developed simple bilayer structure consisting of hole transporting *N*,*N*′-bis(naphthalene-l-yl)-*N*,*N*′-bis(phenyl) benzidine and electron transporting bis(10-hydroxybenzo[h]quinolinato) beryllium layers.

Recently, organic phosphorous compounds have been proposed as electron transport and host layers for low voltage PHOLEDs [8,9]. 4,4′-bis(diphenylphosphine oxide) biphenyl [8] and 2,8-bis(diphenylphosphoryl) dibenzothiophene [9] have been used as electron transporting materials for blue-emitting PHOLEDs. In this paper, we report the low voltage red PHOLEDs with double electron transport layer structure consisting of triphenylphosphine oxide (Ph_3PO) and 4,4′-bis(2,2′-diphenylvinyl)-1,1′-biphenyl (DPVBi) layers. The Ph_3PO, which has been used to form complexes with lanthanide metal ions for using an emitter in OLEDs [10], has a high electron transporting ability [11]. The DPVBi has been widely used as a charge transporting host material in fluorescent OLEDs [12]. In our devices, we used the DPVBi layer as an electron transporting material. By using the Ph_3PO and DPVBi electron transport layers, we demonstrate the red PHOLEDs with operating voltages of 2.8–3.0 V at a luminance of 1000 cd/m^2.

2. Experimental

Red PHOLEDs were fabricated on indium tin oxide (ITO) coated glass substrates. The sheet resistance of ITO film was about 10 Ω/sq. After defining the ITO anode patterns using standard photolithography process, the substrates were cleaned with isopropyl alcohol and deionized water followed by exposing to oxygen plasma. All organic and metal layers were deposited by using a thermal

* Corresponding author. Tel.: +82 41 530 1312; fax: +82 41 530 1722.
E-mail address: dgmoon@sch.ac.kr (D.-G. Moon).

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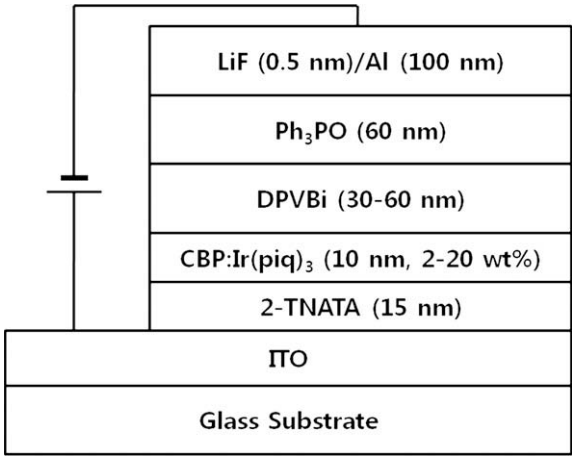

Fig. 1. Schematic diagram of the red PHOLED structure.

evaporation method in a base pressure of about 1×10^{-6} Torr. A 15 nm thick 4,4′,4″-tris[*N*-(2-naphthyl)-*N*-phenyl-amino]-triphenylamine (2-TNATA) layer was deposited on the patterned ITO substrate, and then 4,4′-bis(*N*-carbazolyl)-1,1′-biphenyl (CBP) layer was co-deposited with a red phosphorescent dopant, tris-(1-phenylisoquinolinolato-C^2,*N*) iridium(III) [Ir(piq)$_3$], whose concentration was varied from 2 to 20 wt.%. After then, 30–60 nm thick DPVBi layers were deposited, followed by the deposition of a 60 nm thick Ph_3PO layer. After depositing organic layers, a 0.5 nm thick LiF and a 100 nm thick Al layers were sequentially evaporated through a shadow mask. Fig. 1 shows a schematic diagram of the completed device structure. All the completed devices were encapsulated without exposing to air in a nitrogen atmosphere glove box. Current density–voltage–luminance (*J*–*V*–*L*) characteristics of the devices were measured using computer controlled Keithley 2400 source-measure units and a luminance meter (Minolta LS 100). Electroluminescence (EL) spectra and color coordinates were measured with a spectroradiometer (Minolta CS 1000).

3. Result and discussion

Fig. 2 shows the *J*–*V*–*L* curves of the red-emitting phosphorescent devices. The device structure is ITO/2-TNATA (15 nm)/CBP:Ir(piq)$_3$ (10 nm, 6 wt.%)/DPVBi (30–60 nm)/Ph_3PO (60 nm)/LiF/Al. The devices operate at significantly low voltages. For example, the turn-on voltage for a luminance of 1 cd/m^2 is 2.2 V in the device with a 30 nm thick DPVBi layer. The luminance in this device increases very rapidly with increasing the voltage, being a high luminance of 1000 cd/m^2 at a low voltage of 3.0 V. The driving voltages of our devices are lower than those of the reported red phosphorescent devices [7,13]. For example, Tsuzuki and Tokito [13] demonstrated a red device with an operating voltage of 5 V at a luminance of 1000 cd/m^2, using a bis(2-phenylpyridinato-*N*,$C^{2'}$)iridium(acetylacetonate) as a narrow band gap host for the red phosphorescent Ir(piq)$_3$ guest. The driving voltage is also lower than that of the *pin* structured red PHOLED (~3.5 V at 1000 cd/m^2) reported by Reineke et al. [14]. The low driving voltages in our devices are clearly attributed to the high current densities as shown in Fig. 2. For example, a voltage of 3.2 V is required to achieve a current density of 20 mA/cm^2 in the device with a 30 nm thick DPVBi layer. On the other hand, a 60 nm thick DPVBi device needs about 0.2 V higher voltage (3.4 V) for the same current density. Thus, the results indicate that the voltage drop across the DPVBi layer is small. It also suggests that the DPVBi has a good capability of transporting electrons. In addition, as described in a previous report [11], the required voltages for the same current density were almost independent of Ph_3PO thickness (2.9–3.1 V for 20 mA/cm^2) in the devices with a structure of ITO/2-TNATA/DPVBi/Ph_3PO/LiF/Al. The electrons can be easily transported thorough the Ph_3PO layer and can be easily injected from Ph_3PO layer into the DPVBi layer. Therefore, most of applied voltage is dropped across the 2-TNATA and doped CBP layers rather than the DPVBi and Ph_3PO layers. Since the thicknesses of 2-TNATA and Ir(piq)$_3$ doped CBP layers are 15 and 10 nm, respectively, high electric fields are built in these thin layers, resulting in high current density at low applied voltage.

Fig. 3 shows the *J*–*V*–*L* curves of the red PHOLEDs doped with various concentrations of Ir(piq)$_3$. Although the Ir(piq)$_3$ doped CBP layer is only 10 nm, the current density at the same voltage is higher at higher concentration of Ir(piq)$_3$. For example, the current density at 4 V increases from 76 to 135 mA/cm^2 when the Ir(piq)$_3$ concentration increases from 2 to 20 wt.%, so that the device doped with 20 wt.% Ir(piq)$_3$ exhibits a significantly low driving voltage. The 20 wt.% Ir(piq)$_3$ doped device shows the operating voltages of 2.4 and 2.8 V at 100 and 1000 cd/m^2, respectively. This dependency of the Ir(piq)$_3$ concentration on the current density indicates the direct injection of charge carriers into the guest molecules from adjacent carrier transport layers. The highest occupied molecular orbital (HOMO) levels of 2-TNATA and Ir(piq)$_3$ are 5.1

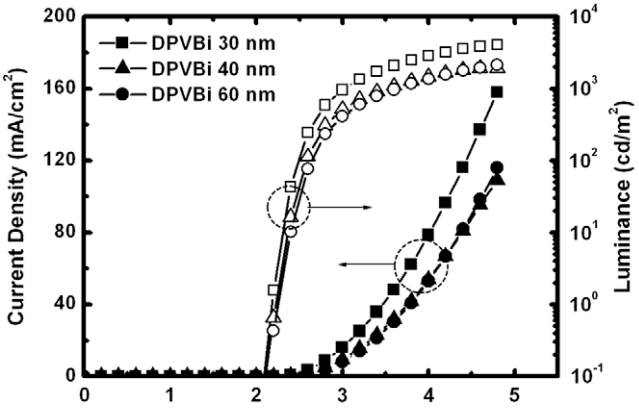

Fig. 2. Current density (solid)–voltage–luminance (open) curves of the red PHOLEDs with different thicknesses of DPVBi layers. Device structure: ITO/2-TNATA (15 nm)/CBP:Ir(piq)$_3$ (10 nm, 6 wt.%)/DPVBi (*x* nm)/Ph_3PO (60 nm)/LiF (0.5 nm)/Al, where *x* = 30, 40, and 60.

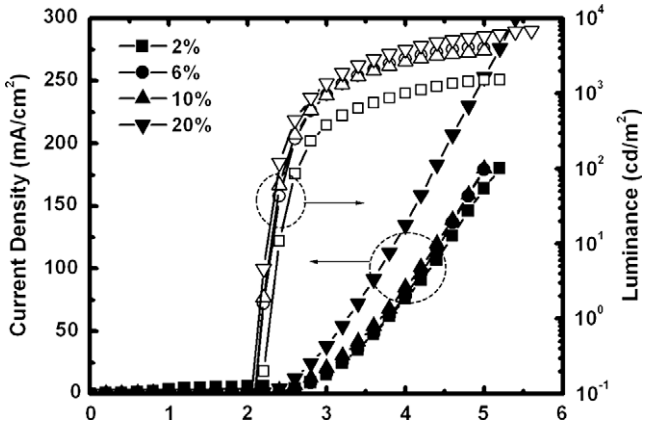

Fig. 3. Current density (solid)–voltage–luminance (open) curves of the red PHOLEDs with different concentrations of Ir(piq)$_3$. Device structure: ITO/2-TNATA (15 nm)/CBP:Ir(piq)$_3$ (10 nm, *x* wt.%)/DPVBi (30 nm)/Ph_3PO (60 nm)/LiF (0.5 nm)/Al, where *x* = 2, 6, 10, and 20.