Improved Efficiency of Organic Light-Emitting Diodes with Doped Transporting Layer

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Abstract

We demonstrate p-doped organic light emitting diodes (OLEDs) comprising tungsten oxide (WO$_3$) and 1,4-bis[N-(1-naphthyl)-N'-phenylamino]-4,4' diamine (NPB). We propose the NPB : WO$_3$ composition functions as a p-doping layer which significantly improves hole injection that leads to the fabrication of 4-(dicyano-methylene)-2-methyl-6-(p-dimethyl aminostyryl)-4H-pyrane (DCM1) based p-doped OLEDs with high efficiency and long lifetime.

1. Introduction

Since the efficient organic light-emitting diodes (OLEDs) have been discovered, there has been considerable interest in developing OLEDs with high efficiency and long lifetime for display applications [1-2]. In order to high efficiency, it is critical to enhance the carrier injection from the electrode to the transporting layer and to increase the transport conductivity [3-4]. Recently, the p-doping hole transporting layer (HTL) for enhancing carrier injection and lowering drive voltages in OLEDs has attracted a lot of attention. The p-doping HTL is typically made by coevaporating the hole transporting material with a strong electron acceptor such as tetrafluro-tetracyanoquinodimethane (F4-TCNQ) [5], or oxidants such as antimony pentachloride (SbCl$_5$) [6], ferric chloride (FeCl$_3$) [7], and iodine [8]. However, using highly volatile F4-TCNQ in thermal evaporation under high vacuum has raised serious concerns over issues of cross contamination, chamber pollution, and the thermal stability. We used tungsten oxide (WO$_3$) as a p-dopant which exhibits good

electric properties [9]. In this study, an inorganic electron acceptor, WO$_3$, was coevaporated with and 1,4-bis[N-(1-naphthyl)-N'-phenylamino]-4,4' diamine (NPB) to form a p-doping HTL in devices with 4-(dicyano-methylene)-2-methyl-6-(p-dimethylaminostyryl)-4H-pyrane (DCM1) emitter.

2. Experimental

For the study of hole injection and transporting ability of NPB:WO$_3$, a series of devices, ITO / NPB : x % WO$_3$ (55 nm) / NPB (10 nm) / tris-(8-hydroxyquinoline) aluminum (Alq$_3$) : DCM1 (30 nm) / 4,7-diphenyl-1,10-phenanthroline (BPhen) (35 nm) / lithium quinolate (Liq) (20 nm) / alumnum (Al) (100 nm), was fabricated. The schematics are depicted as in Fig. 1.
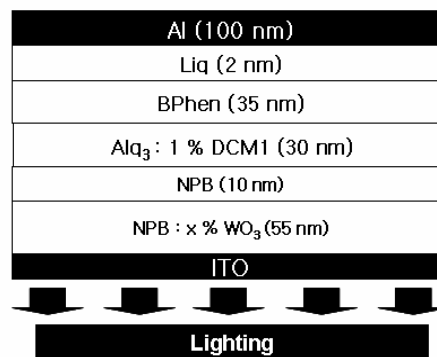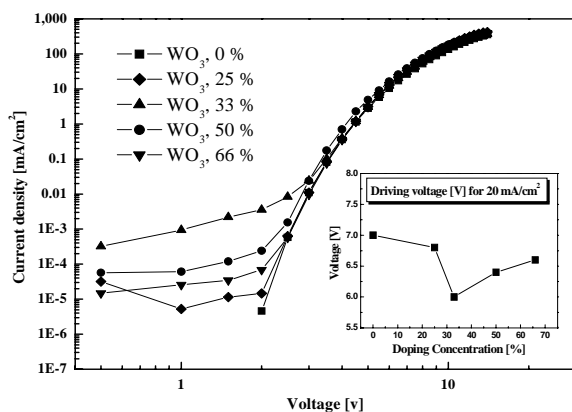

Fig. 1. The structure of OLEDs with 0 %, 25 %, 33 %, 50 % and 66 % WO$_3$ doped NPB films.
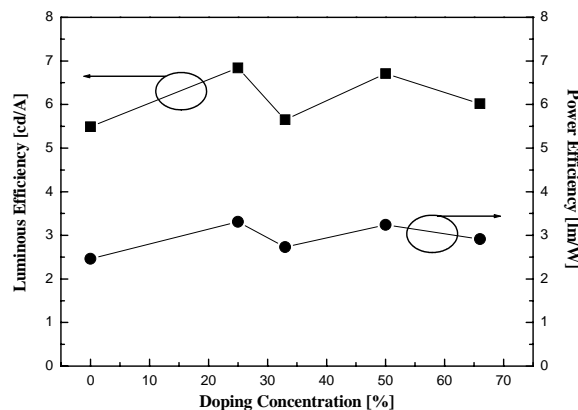
The ITO glass was chemically cleaned using acetone, methanol, distillated water, and isopropyl alcohol. The OLEDs were fabricated by high vacuum (5×10^{-7} Torr) thermal deposition of organic materials onto the surface of an indium tin oxide (ITO, 30 Ω/ $\square$, 80 nm) coated glass substrate. The typical organic deposition rate was 0.1 nm/sec. The active area of the OLEDs was 0.09 cm^2. After fabrication, the current density–voltage (J-V) and degradation characteristics of the OLEDs were measured with a source measure unit (Kiethley 236 and Kiethley 617) and the luminance and CIE chromaticity coordinates of the fabricated devices were measured using a chromameter (MINOLTA CS-100A). All measurements were performed in ambient conditions under a DC voltage bias.

3. Results and discussion

The J-V characteristics at various doping ratios of WO_3 to NPB are shown in Fig. 2. Increasing the WO_3 volume percentage from 0 % to 33 % enhances the current density gradually which is due to the decrease of the resistance and activation energy of the doping layer [10]. However, the current density is reduced slightly as the doping ratio further increases to 50 % and 60 %. This result may be attributed to the effect of carrier quench and generation of defects. The figure inserted in Fig. 2 shows driving voltage of devices with various doping ratio of WO_3 at current density of 20 mA/cm^2. The driving voltage trend is consistent with the current density characteristics. When the

Fig. 3. Luminous efficiency and power efficiency vs doping concentration characteristics at 20 mA/cm^2.

doping ratio increases to 33 %, device has the lowest driving voltage with the high current enhancement. The driving voltages of devices with 0, 25, 33, 50 and 66 % doped WO_3 ratio are 7.0, 6.8, 6.0, 6.4 and 6.6 V. Furthermore, the luminous efficiency and power efficiency characteristics are highly dependent on the doping ratio of WO_3.

The power efficiency of device with a doping ratio of 25 %, 3.3 lm/W (corresponding to the highest luminous efficiency of 6.8 cd/A) at 20 mA/cm^2, is higher than that of these devices with doping ratio of 0, 33, 50 and 66 %, which are 2.5, 2.7, 3.2 and 2.9 lm/W (corresponding to the luminous efficiencies of 5.5, 5.7, 6.7 and 6.0 cd/A) at 20 mA/cm^2, respectively, as
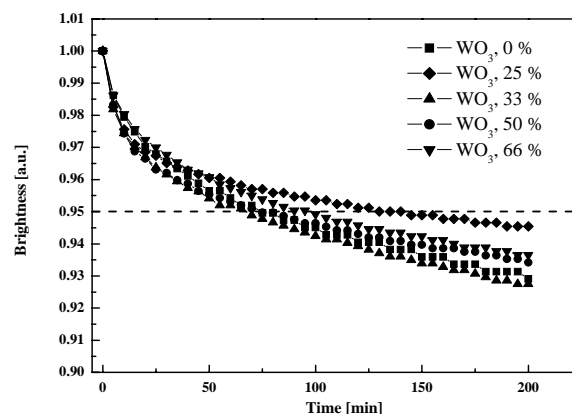

Fig. 2. Current density vs voltage characteristics of devices with various WO_3 doping concentration. Inset: Voltage vs doping concentration characteristics at 20 mA/cm^2.

Fig. 4. Degradation curves of devices driven at room temperature under initial luminance of 500 cd/m^2.

shown in Fig. 3. The efficiencies characteristics of the devices is not consistent with current density and driving voltage trends. It is because that the efficiencies depend on the balance of the hole and electron. That is, current density adjusted by WO_3 doping changes the efficiency of devices. The lifetimes of the encapsulated devices were examined under the initial luminance of 500 cd/m^2, and the result is shown in Fig. 4. The 25 % WO_3 doped device show the longest lifetime with t_{95} exceeding about 125 min, while both 0 % and 33 % WO_3 doped devices indicate the shortest lifetime with t_{95} of about 65 min. That is thought that the lifetime results are affected by efficiency characteristics of devices. The quenchers in device are produced by hole and electron not to participate in emission. Thus, balanced hole electron lead to high efficiency and long lifetime.
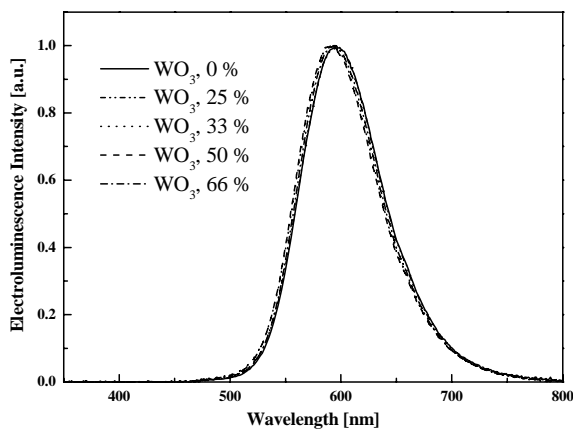
Fig. 5. EL spectra of devices driven at applied voltage of 12 V.

Fig. 5 shows the EL spectra of devices driven at applied voltage of 12 V. EL maxima of all devices appear at about 595 nm, which is orange emission due to DCM1. The emission color is regardless of WO_3 doping ratio in devices.

4. Summary

Hole injection is considered to be one of the most important factors that dictate the driving voltage and power efficiency of OLEDs. In this letter, we use WO_3 as inorganic metal oxide for the electron-acceptor dopant in HTL. We have demonstrated the improved lifetime, driving voltage (6 V for 20 mA/cm^2) in OLED with 33 % WO_3 doped NPB film. If p-i-n

OLEDs were fabricated to balance of recombined hole and electron, the efficiency as well as lifetime and driving voltage will be much more improved at once.

5. References

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