

Efficiency Improvement of Organic Light-emitting Diodes depending on Thickness of Hole Injection Materials

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In the device structure of ITO/hole injection layer/N, N'-biphenyl-N, N'-bis-(1-naphenyl)-[1,1'-biphenyl]4,4'-diamine(NPB)/tris(8-hydroxyquinoline) aluminum(Alq₃)/Al, we investigated an effect of hole-injection materials (PTFE, PVK) on the electrical characteristics and efficiency of organic light-emitting diodes. A thermal evaporation was performed to make a thickness of NPB layer with a evaporation rate of 0.5 ~ 1.0 Å/s in a base pressure of 5×10⁻⁶ Torr. We measured current-voltage characteristics and efficiency with a thickness variation of hole-injection layer. The PTFE and PVK hole-injection layer improve a performance of the device in several aspects, such as good mechanical junction, reducing the operating voltage and energy band adjustment. Compared with the devices without a hole-injection layer, we have obtained that an optimal thickness of NPB was 20 nm in the device structure of ITO/NPB/Alq₃/Al. And using the PTFE or PVK hole-injection layer, the external quantum efficiencies of the devices were improved by 24.5 % and 51.3 %, respectively.

Keywords : Hole injection layer(PTFE, PVK), OLEDs, External quantum efficiency

1. INTRODUCTION

Organic light-emitting diodes (OLEDs) have been widely studied for next generation flat panel display because of possible application in display, low-operating voltage, low-power consumption, low-cost, self-emission and capability of multicolor emission by the selection of emissive materials[1-3]. The first report on organic electroluminescence was published by Helfrich and Schneider in 1965 with anthracene[4,5]. But there was not much progress in 1960's and 1970's because of limited size, difficulty of single crystal growth and high driving voltage. In 1990, Friend *et. al.*, in Cambridge University reported first green light-emitting polymer diodes using poly phenylenevinylene (PPV)[6]. Since then, lots of progress has been done to obtain highly

efficient and stable light-emitting diodes for the last twenty years[7-14].

In this paper, in order to enhance the performance of OLEDs, some organic materials are adopted for hole-injection layer inserted between indium-tin-oxide (ITO) and the emissive layer. The N,N' - biphenyl-N, N'-bis-(1-naphenyl)-[1,1'-biphenyl] 4, 4'-diamine (NPB)[15] was used as a hole-transport layer, and the polytetrafluoroethylene (PTFE)[16] and polyvinyl-carbazole (PVK)[17] were used as a hole injection layer. In the ITO/hole injection layer/NPB/tris(8-hydroxy-quinoline) aluminum(Alq₃)/Al device, we have studied the effects of PTFE and PVK hole injection layer in OLEDs based on Alq₃ thin films by investigating current-voltage characteristics, luminance-voltage characteristics, luminous efficiency and external quantum efficiency.

2. EXPERIMENTAL

In our experiments, we used NPB as a hole-transport and Alq₃ as an electron transport and emissive material. Two device structures were made: one is ITO/NPB/Alq₃/Al as a reference, while the other is ITO/hole injection layer/Alq₃/Al to be used to investigate the effects of hole injection layer. The ITO glass, having a sheet resistance of 15 Ω/ and thickness of 170 nm, was received from Samsung Corning. A 5 mm wide ITO strip line was made by selective etching using a vapor of solution by mixing with hydrochloric acid (HCl) and nitric acid (HNO₃) at a volume ratio of 3:1 for 20 minutes at room temperature.

The distance between the ITO and etch ant was about 2 cm. Then, the patterned ITO glass was cleaned ultrasonically in chloroform for 20 minutes at 50 °C, after

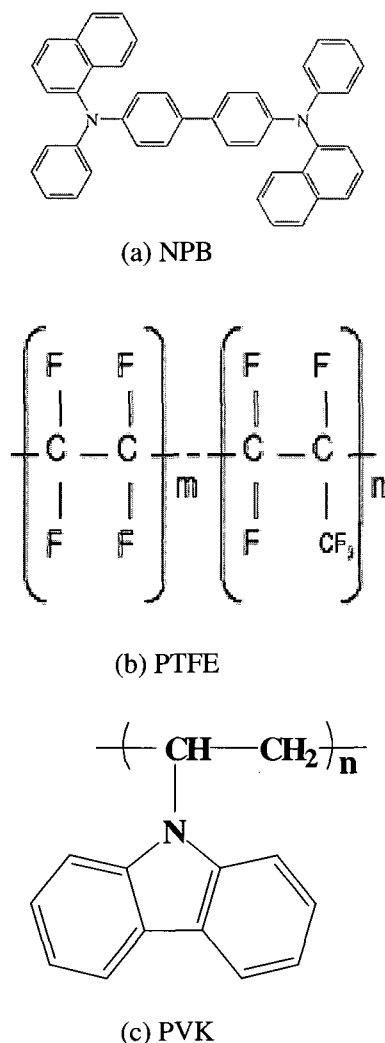


Fig. 1. Molecular structures of hole-transport (a) and hole-injection layer (b), (c).

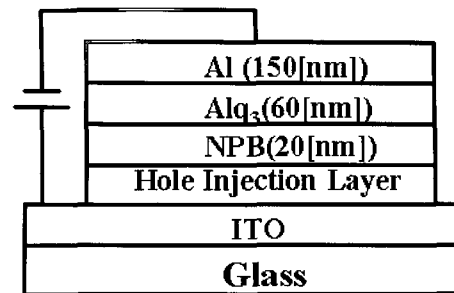


Fig. 2. Device structure of OLEDs.

which the ITO glass was heated at 80 °C for 1 hour in a solution made with second distilled deionized water, ammonia water and hydrogen peroxide at a volume ratio of 5:1:1. We sonicated the substrate again in chloroform solution for 20 minutes at 50 °C, in acetone for 20 minutes at 50 °C and in deionized water for 20 minutes at 50 °C. After sonication, the substrate was dried with N₂ gas stream and was stored under vacuum.

Figure 1(a), (b), and (c) show molecular structures of NPB used as a hole-transport material and PTFE, PVK used as a hole injection layer. Figure 2 shows a device structure of ITO/hole injection layer/NPB/Alq₃/Al to investigate the influence of hole injection layer. Also, we investigated electrical characteristics of organic light-emitting diodes with the thickness variation of hole injection layer. The film thickness of PTFE was made to be 0.5, 1.0, 1.5, and 2.0 nm, and that of PVK to be 10.5, 11.0, 11.5, 12.5, and 13.5 nm.

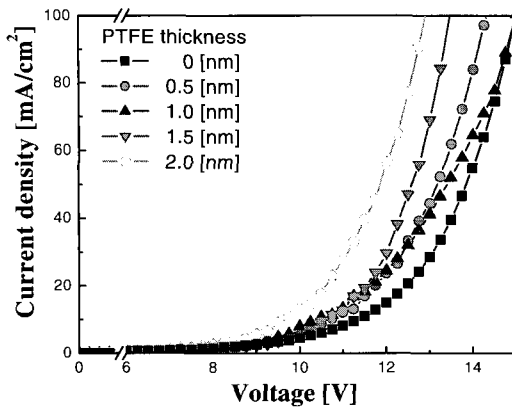
Luminous efficiency is useful for the display application. It is given by $\eta = \pi L/JV$ [lm/W], where L [cd/m²] is the out coming luminance that is measured normal to the emitting surface, J [A/m²] is the current density, and V [V] is the applied voltage.

3. RESULTS AND DISCUSSION

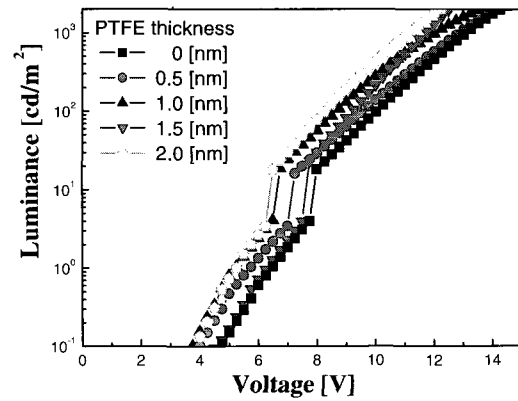
3.1 Characteristics of PTFE hole injection layer

Figure 3 shows the current density-voltage and luminance-voltage characteristics of ITO/PTFE/NPB(20 nm)/Alq₃/Al device, and with a thickness variation of PTFE layer from 0.5 to 2.0 nm. As the thickness of PTFE layer increases, the current density and the luminance also increase as well. When the thickness of PTFE layer is 1.0 nm, the efficiency is higher than the others.

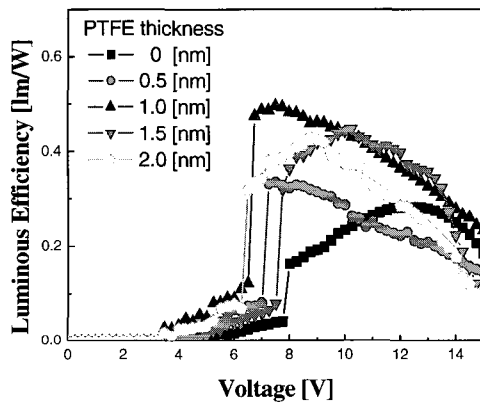
The efficiency strongly depends on charge balance. The PTFE layer helps more holes to be injected into the emissive layer. By inserting thin layer of insulating material such as PTFE, it acts either as a reducing a work function of the hole transport layer. It is thought that the PTFE layer controls a hole injection and recombination rate. Fig. 3(d) shows the external quantum efficiency of the device.



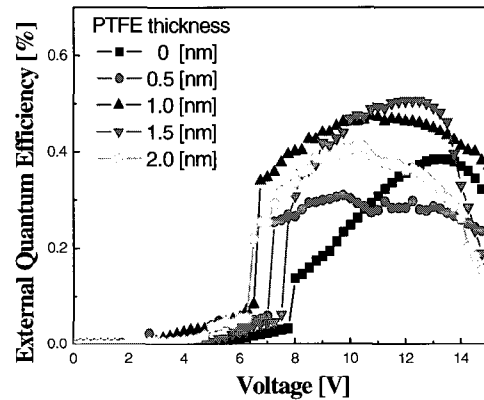
(a) Current density-Voltage characteristics



(b) Luminance-Voltage characteristics



(c) Luminous efficiency-Voltage characteristics



(d) External quantum efficiency-Voltage characteristics

Fig. 3. I-V-L- η of hole injection layer (PTFE)

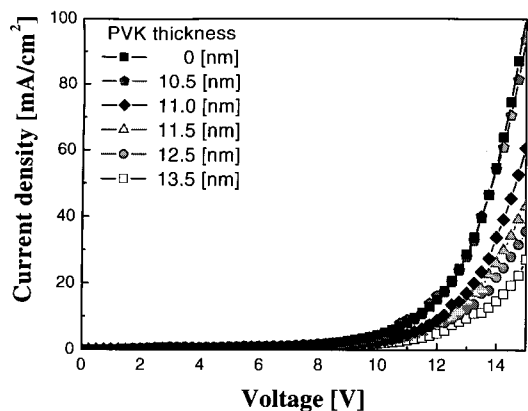
The external quantum efficiency begins to increase near 6 V and becomes maximum value near 13 V.

It was found that the device without the PTFE layer shows the external quantum efficiency of 0.38 %. However, with a use of 1.0 nm thickness of PTFE layer. The external quantum efficiency becomes 0.47 %. The improvement of the efficiency is about 24.5 % with a use of PTFE layer. As the thickness of PTFE layer increases up to 1.0 nm, the efficiency increases as well. However, when the thickness of PTFE layer becomes 2.0 nm, the efficiency decreases. When the PTFE layer increases further, it acts like a resistive layer for hole-injection layer.

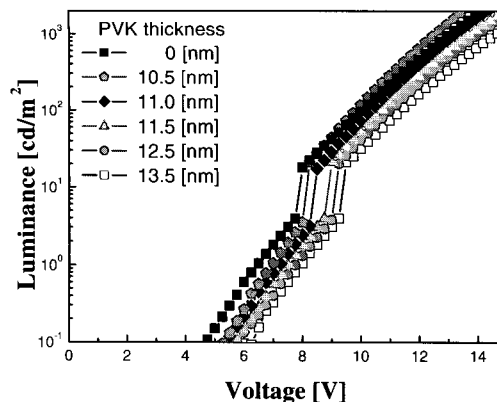
3.2 Characteristics of PVK hole injection layer

Figure 4 shows typical nonlinear current-voltage characteristics shows the current-voltage and luminance-voltage characteristics of ITO/PVK/NPB (20 nm)/Alq3 (60 nm)/Al with a thickness variation of PVK from 10.5 to 13.5 nm. As the voltage increases above 5 V, the

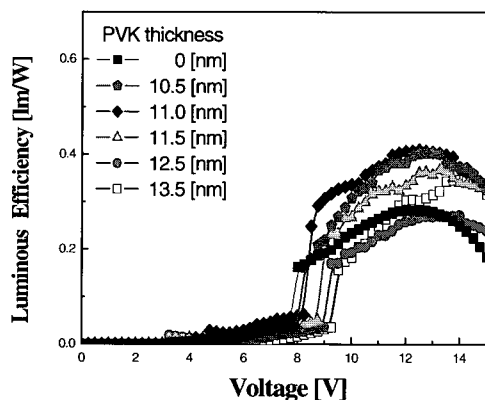
current density and the luminance start to increase. As the thickness of PVK layer increases, the current density and corresponding luminance decrease. The luminous efficiency and external quantum efficiency increase gradually for then layer thickness and decrease for thicker one. The efficient hole injection is expected to enhance the efficiency in the OLEDs. It is thought that a recombination rate in the emissive layer increases as the PVK layer thickness increases because the PVK layer prevents the hole injection properly. Figure 4(d) shows a quantum efficiency of the devices. The external quantum efficiency starts to increase from 8 V and reaches a maximum near 13 V. In the case of device without hole injection layer, the maximum efficiency is about 0.38 %. On the other hand, the device having a 11.0 nm thick PVK layer gives an efficiency of about maximum 0.575 %. With a use of PVK layer, there is an enhancement of efficiency by 51.3 %. But, it shows that the efficiency is reduced if the thickness of PVK increases to 13.5 nm.



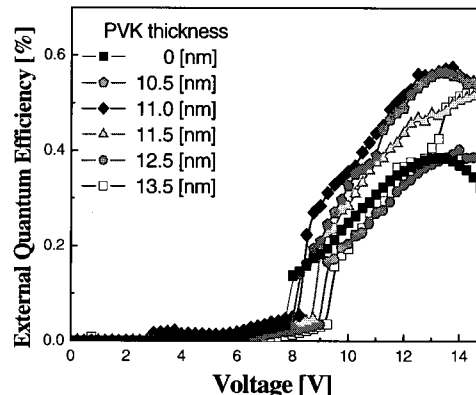
(a) Current density-Voltage characteristics



(b) Luminance-Voltage characteristics



(c) Luminous efficiency-Voltage characteristics



(d) External quantum efficiency-Voltage characteristics

Fig. 4. I-V-L- η of hole injection layer(PVK).

Thus, we confirmed that optimum luminance and external quantum efficiency could be obtained by using a proper thickness of the PVK layer.

4. CONCLUSION

To investigate the effect of the hole-injection materials on the electrical characteristics, we fabricated ITO/hole injection layer/NPB/Alq₃/Al device structure. When introduced into OLEDs as a hole injection layer, PTFE or PVK layer can efficiently enhance the hole injection in order to achieve a hole-electron balance in the OLEDs. It was found that the device without the PTFE layer showed the external quantum efficiency of 0.38 % while the device with 1.0 nm PTFE layer showed the external quantum efficiency of 0.47 % resulting in 24.5 % efficiency improvement. When a PVK layer was used, it was turned out that the effect was more significant. The device without the PVK layer showed

0.38 % external quantum efficiency and the device with 11.0 nm PVK layer showed the external quantum efficiency of 0.575 %, which results in 51.3 % improvement in efficiency.

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