

Improvement of Performance and Color Stability of Solution-Processed OLED by Controlling Recombination Zone

Thi Na Le and Min Chul Suh*

Email: mcsuh@khu.ac.kr

Department of Information Display, Kyung Hee University, Seoul 02447, Republic of Korea

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ABSTRACT

The recombination region is found on the hole transport layer/emitting layer side when using a relatively high electron mobility host material. By the way, in this case, the electroluminescence spectrum becomes broader as the driving voltage increases. High field-dependence for electron mobility of host material and electric field-induced quenching of excited state would be responsible for a color instability. On the contrary, the well-designed device utilizing host material with high hole mobility resulted in well-controlled recombination zone which is far away from the critical interface mixing zone. With this device, we obtain improved performance as well as much improved color stability.

1 INTRODUCTION

Organic light emitting devices (OLEDs) have attracted much attention because of their superior characteristics such as vivid colors, thin form factor, flexibility, etc. However, still, most of OLED devices are being prepared by vacuum evaporation technology that has to waste a lot of material. Thus, a development of solution processed OLED manufactured by ink-jet printing technology is becoming very important to overcome this problem. However, for solution processed OLED, we face a situation where the interface must be mixed during consecutive wet processes. Of course, crosslinking techniques are being used to prevent interface mixing, but some of the unreacted monomers are easily mixed with solutions that are continuously deposited on the top of previously solution processed unit layer. However, we couldn't conclude that interface mixing would adversely affect the device characteristics unconditionally or not because we could obtain much better device characteristics if the interface mixed seriously sometimes.^[1]

Therefore, in particular, we tried to control the RZ (recombination zone) by forming the material of the host for EML formation into a hole type or electron type host, and to find out how the characteristics and stability of the device fabricated by the solution process in each case are different.

2 EXPERIMENT

The solution-processed OLED devices were fabricated with 150 nm thick patterned indium-tin oxide

(ITO) glasses covered by a bank layer with an open emission area of 4 mm². The ITO glasses were cleaned in acetone and isopropyl alcohol with a sonication process and were rinsed in deionized water. Then, the ITO glass substrates were treated in UV-ozone to eliminate all organic impurities during the previous fabrication processes. PEDOT:PSS (CH8000) was spin-coated as a hole injection layer (HIL) on ITO glass in an ambient condition and was annealed at 165 °C for 20 min in an inert atmosphere. Subsequently, hole transport layer (HTL, PHS03:Poly-TPD (20wt%)) dissolved in toluene was spin-coated as a hole transport layer and was crosslinked by using a standard process. For preparation of green emitting layer (EML), (2,2',2''-(1,3,5-phenylene)tris(1-phenyl-1H-benzimidazole)) (TPBi) and APPG087 materials used as an electron-type host and hole-type host. And, they were dissolved in methanol and toluene, respectively, to provide a 1 wt% solution. Green emitting layer (EML) was formed and dried at 100 °C for 10 min. All solution processes were performed in a nitrogen atmosphere at room temperature, except for the PEDOT:PSS. After spin coating with green EML, TPBi as an exciton blocking layer (EBL), LG201:LiQ (1:1) as an electron transport layer (ETL) were thermally deposited in a vacuum condition under 10⁻⁷ Torr with 0.5 Å/s. Then, lithium quinolate (LiQ) and aluminum (Al) were deposited successively with 0.3 Å/s and 3 Å/s, respectively.

3 RESULTS AND

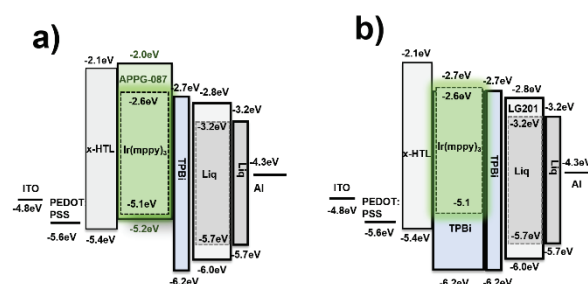


Fig. 1 Device structure of a) **Device A** with APPG087 as host material, b) **Device B** with TPBi as host material.

To compare the device performances with different RZ position we prepared the two different solution processed OLEDs as follows:

Device A: ITO/ PEDOT:PSS (40nm)/ xHTL (17nm)/ APPG087:Ir(mppy)₃ (10%,30nm)/ TPBi (5nm)/ LG201:Liq (1:1, 45nm)/ Liq (1 nm)/ Al (100 nm).

Device B: ITO/ PEDOT:PSS (40nm)/ xHTL (17nm)/ TPBi:Ir(mppy) (10%, 30 nm)/ TPBi (5nm)/ LG201:Liq (1:1, 45nm)/ Liq (1 nm)/ Al (100 nm).

Fig. 1 shows the energy band diagram of devices. A thin layer of TPBi was inserted between EML and ETL in the two devices to serve as EBL (exciton blocking layer).

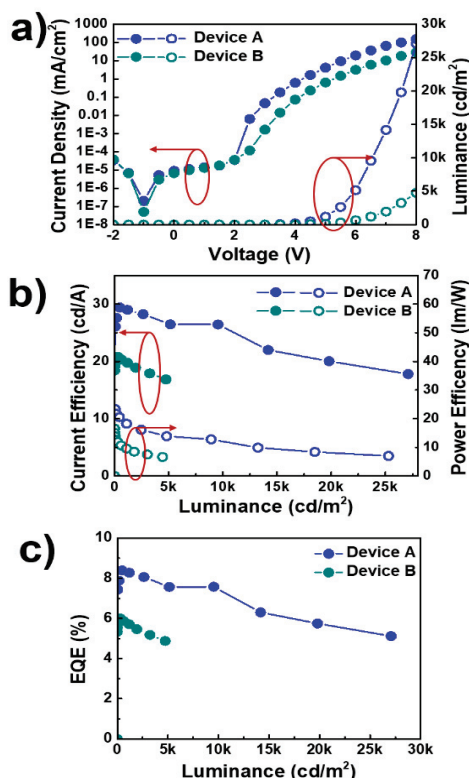


Fig. 2 Performance characteristics of device with APPB087 (**Device A**) and TPBi (**Device B**) as host material. a) Current density-voltage-luminance, b) Current and power efficiency-luminance, c) External quantum efficiency.

Device characteristics were shown in **Fig. 2** and also summarized in **Table 1**. The operating voltages at which the luminance was 1000 cd m⁻² were 4.7V and 6.3V for **Device A** and **Device B**, respectively. **Device A** also exhibits much better current injection, operating voltage and turn on voltage compared to **Device B** (**Fig. 2a**). The current and power efficiency (**Fig. 2b**) at a constant luminance of 1000 cdm⁻² were 29.2 cdA⁻¹ and 18.5 lmW⁻¹ for **Device A**, 20.0 cdA⁻¹ and 10 lmW⁻¹ for **Device B**, respectively. The maximum current and power efficiencies were 29.4 cdA⁻¹ and 23.4 lmW⁻¹ for **Device A**, 20.8 cdA⁻¹ and 16.5 lmW⁻¹ for **Device B**, respectively. **Device A** also exhibits a much higher luminance, which are 27,086 cd/m² and 4,742 cd/m² for **Device A** and **Device B**, respectively.

Table 1: Summary of device characteristics

Device	V _{on} ^(a) /V _{op} ^(b) [V]	CE ^(c) /PE ^(d) /EQE ^(e) [cdA ⁻¹ /lmW ⁻¹ /%]		CIE ^(f)
		Max	at 1,000cdm ⁻²	
Device A	2.8/4.7	29.4/23.4/8.4	29.2/18.5/8.3	(0.302, 0.622)
Device B	3.4/6.3	20.8/16.5/6.0	20.0/10.0/5.8	(0.310, 0.617)

^(a)V_{on}: turn on voltage, measured at 1cdm⁻² ^(b)V_{op}: operating voltage measured at 1,000cdm⁻² ^(c)CE: current efficiency ^(d)PE: power efficiency ^(e)EQE: external quantum efficiency ^(f)CIE: Commission International de L'Eclairage.

The lower performance of **Device B** was probably resulted from the occurrence of recombination zone at interface mixing zone. Especially, we found that the RZ was gradually shifted toward this critical interface during increasing the operation voltage. As shown in **Fig. 3b**, relatively low electron injection barrier between EBL and EML can cause the electrons to be easily injected and transported through EML to be accumulated at HTL/EML interface. Meanwhile, a high hole injection barrier at HTL/EML and low hole mobility of HTL would be also reasons for formation of RZ near/at the HTL/EML interface. However, a relatively low electron mobility of TPBi ($\mu_e \sim 10^{-5} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) causes field dependent spectral broadening effect during increase in operation voltage as aforementioned as shown in **Fig. 4**. In other words, the RZ is further shifted toward HTL/EML interface^[2,3] as voltage increases, due to a relatively high field-dependence of electron mobility in TPBi. This result in – ‘pushing of exciton recombination zone toward an interface mixing zone (HTL/EML) where high charge density accumulated can cause an electric field-induced quenching of excited state resulting in the lower performance and an instability of electroluminescence spectrum.

On the other hand, in **Device A**, a small energy barrier for hole injection into EML from HTL and high hole mobility of host materials could confine the exciton inside EML and far away from interface mixing zone as shown in **Fig. 3**. As a result, **Device A** gave much better performances with high color stability.

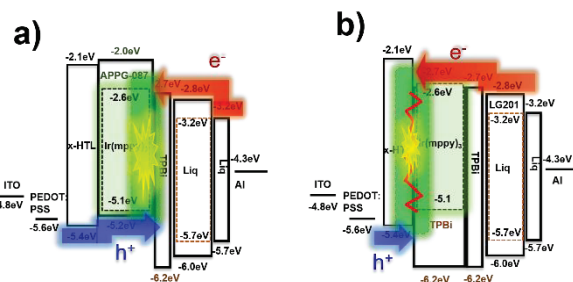


Fig. 3 Illustration of recombination zone position.

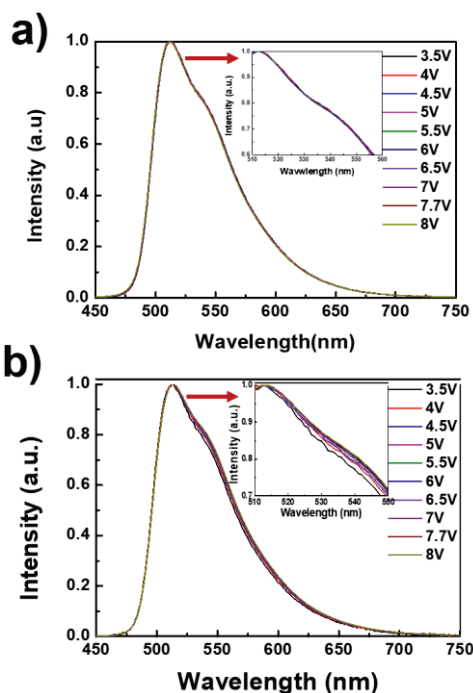


Fig. 4 a) Shift of electroluminescence spectrum with applied voltages of a) device with APPG087 as host (**Device A**) and b) device with TPBi as host material (**Device B**).

4 CONCLUSIONS

We found that the characteristics of solution processed OLEDs differ greatly depending on the location of RZ. In particular, when interface mixing of solution processed OLED has a positive and negative effect on device characteristics, two devices with completely different RZ positions were made to compare devices.

In conclusion, when using a hole type host (APPG087), the interface mixing of HTL and EML does not impair the device characteristics, but when using an electron type host (TPBi), the RZ coincides with the interface mixing zone, which seriously impairs the device characteristics.

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