

Significant Lifetime Enhancement of Organic Light Emitting Diodes by Removing Residual Water during Device Fabrication

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Abstract

We demonstrate that a significant enhancement of operational stability of organic light emitting diodes (OLEDs) can be realized by fabricating the devices under ultra-high vacuum (UHV) conditions in the region of 10^{-10} ~ 10^{-11} Torr. UHV condition is achieved by utilizing non-evaporable getter pumps together with regular turbo molecular pumps in OLED deposition chamber. The short-term and long-term stability of OLEDs is significantly prolonged due to the removal of detrimental gases, especially water. We suggest that ultra-clean fabrication conditions are indispensable to uncover the true intrinsic degradation mode of OLEDs.

1. Introduction

The study on effect of moisture on OLEDs stability has been reported by several groups with different approaches [1–9]. These studies consistently reported that water was the main agent in devices degradation, which takes place by the reaction of tris(8-hydroxyquinolinolato) aluminum (III) (Alq₃) molecules with water molecules [10]. We have reported that a decrease of the amount of residual water by lowering the pressure of an evaporation chamber to ultra-high vacuum (UHV) pressure to the order of 10^{-9} Torr could provide better device stability as compared to that of device fabricated at HV pressure [1,2]. In line with these results, if OLEDs are fabricated under lower region of UHV conditions ($<10^{-10}$ Torr), we should be able to eliminate the extrinsic degradation mode so that we can investigate the true intrinsic degradation mode of OLEDs. In this work, to further evacuating the residual gas in the evaporation chamber, non-evaporable getter pumps (NEGPs) based on the quaternary getter alloy of Ti–Zr–V–Al [11] were added along with turbo molecular pumps (TMPs).

2. Experimental Section

A OLED was fabricated on an indium-tin-oxide (ITO)-coated substrate, on which the multi-layers consist of a molybdenum trioxide layer (0.75 nm), a 90 nm-thick N, N'-di-[(1-naphthyleyl)-N,N'-diphenyl]-1,1'-biphenyl-4,4'-diamine (α -NPD) and a 70 nm-thick Alq₃, lithium fluoride (LiF) (1 nm) and an aluminum layer (Al) (100 nm). Two NEG pumps (pumping speeds of 400 l/s and 1000 l/s) were installed right next to a substrate holder in the evaporation chamber. The OLEDs were fabricated with activating the NEG pumps and without activating the NEG pumps. The luminance – time characteristics of the OLEDs were measured at a constant dc current density of 50 mA/cm².

3. Results and Discussion

Figure 1 shows the current density-voltage-initial luminance (J–V–L) characteristics of the devices fabricated with TMPs or TMPs and NEGPs. The similar device characteristics suggest that adding NEGPs does not affect the initial device characteristics.

Figure 2 demonstrates changes of the luminance and the voltage as a function of an operation time for the OLEDs. The initial operation voltages are consistent (12.4 V) corresponding to an initial luminance of approximately 1700 cd·m⁻² for all devices. Despite the same increase in the operational voltage, the time to reach to 90% of the initial luminance (LT90=136 h) in the device fabricated with TMPs and NEGPs was five times longer than that (LT90=25 h) in the device fabricated with TMPs.

Table 1 summarizes the residual gas components in the initial condition of the evaporation chamber evacuated by either TMPs, or TMPs and NEGPs. It appears that NEGPs effectively evacuate large amount of each detrimental gas from the chamber. By using NEGPs, the partial pressure signals acquired by RGA of H₂O (m/z=18), N₂/CO+ (m/z=28) and CO₂+ (m/z=44) reduced by the 99%, 96% and 95%, respectively, compared to the case using only TMPs. The effect of NEGPs on evacuation of the residual gasses was not only proven at the initial chamber condition but also during OLED device fabrication. To obtain more insights of the enhanced device stability, we calculated the number of water molecules (S) striking the substrates surface in a unit area and time when fabricated with a system evacuated by either TMPs and NEGPs or TMPs [12].

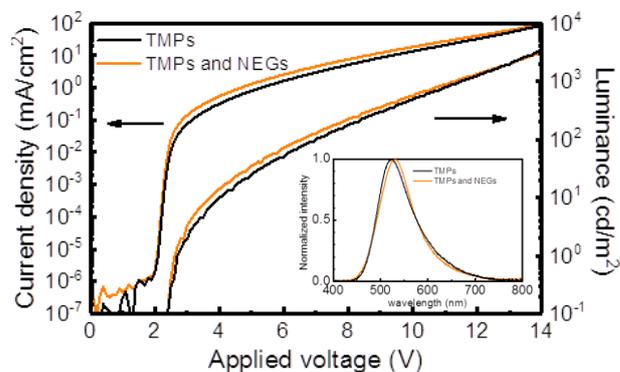


Fig. 1. Current density-voltage-initial luminance characteristics of a device fabricated with TMPs (black) and a device fabricated with TMPs and NEGPs (orange). Inset shows EL spectra of the devices.

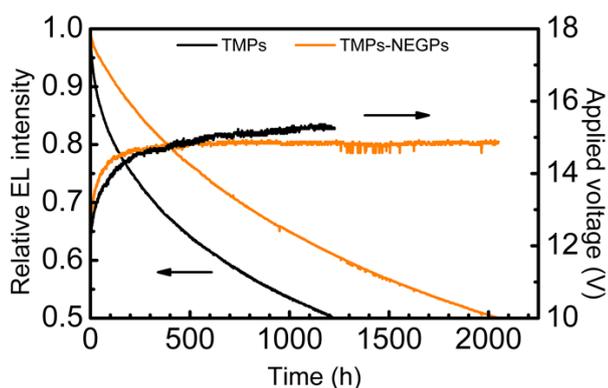


Fig. 2. Luminance/initial luminance–time–operation voltage characteristics of a device fabricated with TMPs, and a device with TMPs and NEGPs.

$$S = \frac{N_A P 10^{-4}}{\sqrt{2\pi k_B M T}}$$

where N_A is the Avogadro number, P is the partial pressure of gas, k_B is the Boltzmann constant, M is the molecular mass of the gas, and T is the temperature. With a density of $1.31 \text{ g}\cdot\text{cm}^{-3}$ and molecular mass of $459.43 \text{ g}\cdot\text{mol}^{-1}$ for Alq_3 , the S of Alq_3 at evaporation rate of $0.1 \text{ nm}\cdot\text{s}^{-1}$ corresponds to $1.70 \times 10^{13} \text{ s}^{-1}\cdot\text{cm}^{-2}$. Similarly, α -NPD molecules ($1.19 \text{ g}\cdot\text{cm}^{-3}$, $588.74 \text{ g}\cdot\text{mol}^{-1}$) [13] strike to the substrate with $1.22 \times 10^{13} \text{ s}^{-1}\cdot\text{cm}^{-2}$. With TMPs and NEGPs, the number of water molecules stroke the substrate ($S=1.18 \times 10^{11} \text{ s}^{-1}\cdot\text{cm}^{-2}$) was about 100 times smaller than those of α -NPD and Alq_3 molecules. In case of the device with TMPs, the number of water molecules stroke the substrate ($S=2.97 \times 10^{12} \text{ s}^{-1}\cdot\text{cm}^{-2}$) was only 4 times smaller than that of α -NPD and Alq_3 molecules. Since the decomposed α -NPD molecules are known to act as an initial quencher [14], the decomposition of α -NPD molecules with the reaction of water molecules would be detrimental to device stability. For the long-term stability of the device (time to reach to 50% of an initial luminance, LT50), the device fabricated with TMPs and NEGPs (LT50=2051 h) was two times longer than that (LT50=1250 h) of the device fabricated with TMPs. It was reported that the reaction of Alq_3 molecules with water molecules would be detrimental to long-term device lifetime [1]. It is clear that the number of water molecules stacking with Alq_3 molecules significantly decrease in the device fabricated with TMPs and NEGPs ($\text{H}_2\text{O}:\text{Alq}_3=1:144$), compared to that ($\text{H}_2\text{O}:\text{Alq}_3=1:6$) of the device fabricated with TMPs.

Surprisingly, we observed almost no change in operation voltage (V driving=14.83 V) in the device fabricated with TMPs and NEGPs after the 500 h operation while the luminance kept decreasing gradually (Fig. 2). This is a clear evidence that the voltage increase no longer relate to either short-term or long-term degradation of the Alq_3 -based OLED. This luminance decay is probably due to luminescent quenching created by the device operation [15].

4. Conclusions

We have demonstrated five times enhancement in the

Table 1. Residual gas components during the device fabrication.

Vacuum Pump	Stability (h)		Partial pressure (Torr)		
	LT90	LT50	H_2O^+ (m/z=18)	N_2^+ or CO^+ (m/z=28)	CO_2^+ (m/z=44)
TMP	25	1,250	6.2×10^{-9}	1.6×10^{-9}	1.3×10^{-9}
TMP-NEG	136	2,051	2.4×10^{-10}	5.5×10^{-11}	8.1×10^{-11}

short-term stability and two times enhancement in long-term stability of Alq_3 -based OLEDs by lowering the residual gas components in the evaporation chamber by using NEGPs in combination with TMPs. We results clearly suggests that the increase in the voltage does not represent either short-term or long-term stability.

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