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Won-Jong Kim [a], Hyun-Min Choi [a], Tae-Wan Kim [b] & Jin-Woong Hong [a]

[a] Department of Electrical Engineering, Kwangwoon University, Seoul, Korea

[b] Department of Physics, Hongik University, Seoul, Korea

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Electrical Conductive Mechanism of Organic Light-Emitting Diodes Depending on the Surface Roughness of Alq_3 Layer

WON-JONG KIM,[1] HYUN-MIN CHOI,[1] TAE-WAN KIM,[2] AND JIN-WOONG HONG[1]

[1]Department of Electrical Engineering, Kwangwoon University, Seoul, Korea
[2]Department of Physics, Hongik University, Seoul, Korea

In a single-layered structure of ITO/Alq_3/Al device, electrical conductive mechanism of organic light-emitting diodes were studied depending on the surface roughness of tris(8-hydroxyquinoline)aluminum (Alq_3) layer. Several different sizes of holes on boat and several different deposition rates were employed in evaporating the organic materials. It was found that the hole size of the crucible boat and the evaporation rate affects the surface roughness of Alq_3 layer as well as the performance of the device. When the hole size of crucible boat and the deposition rate of Alq_3 are 1.0 mm and 2.5 Å/s, respectively, an average surface roughness of the Alq_3 layer is lower and the efficiency of the device is higher than the ones made with other conditions. From the analysis of current density-luminance-voltage characteristics of the device, it was able to identify four conductive mechanisms depending on the voltage region. It was found that a coefficient β_{ST} in Schottky region is 3.28×10^{-24}, a coefficient β_{PF} in Poole-Frenkel region is 6.33×10^{-24}, and a potential barrier φ_{FN} in Fowler-Nordheim region is 0.32 eV.

Keywords Alq_3; conductive mechanism; electrical properties; negative resistance; organic light-emitting diodes; surface roughness

1. Introduction

With the acceleration of information technology by the development of info-communication in the 21st century, there is a trend that the importance of a display gradually transfers to a high contrast display [1,2]. One of the new displays satisfying this trend is organic light-emitting diodes (OLEDs). The OLEDs is a device that electrons and holes injected from the electrode generate light by recombination in organic thin film. Since the OLEDs are self-emissive, it has a wider view angle, high efficiency, and a faster response compared to those of LCD. Also, it does not need a backlight, so that the OLEDs can be thin and light, and still have the advantage in power consumption [3–5]. However, there are still some problems to

Address correspondence to Jin-Woong Hong, Department of Electrical Engineering Kwangwoon University, Wolgye-Dong, Nowon-Gu, Seoul 139-701, Korea (Republic of Korea). Tel.: 82-2-940-5145; Fax: 82-2-915-4630; E-mail: ealab@kw.ac.kr

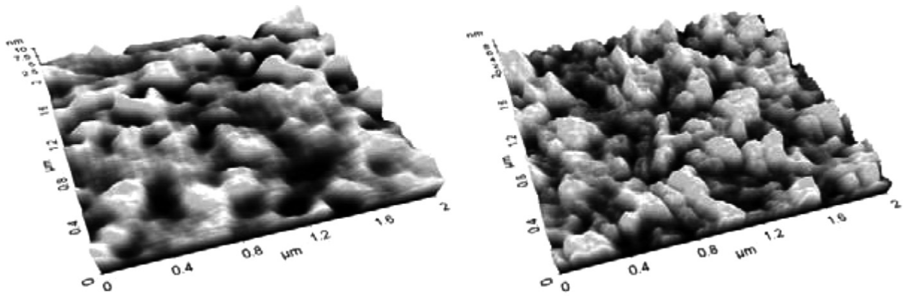
overcome in organic light-emitting diodes. Active studies are continuing to solve problems such as charge injection, transport, and materials, surface roughness, etc [6–8]. In this paper, electrical properties of the organic light-emitting diodes depending on the surface roughness of Alq_3 are presented. A special type of self-made crucible boat was made out of stainless steel rather than using conventional boat for an evaporation of organic materials to improve the efficiency of the device. Self-made crucible boat is cylindrically shaped and has a small hole on top of it. After finding an optimum hole size for evaporation of Alq_3, we studied the electrical mechanism in the device.

2. Experimental

We have used S. Co's ITO (indium-tin-oxide), having a sheet resistance of 15Ω/sq and a thickness of 170 nm as an anode. The ITO substrate was patterned as follows. A 5 mm wide ITO electrode was made by etching it using a vapor of solution made by mixing hydrochloric acid (HCl) and nitric acid (HNO_3) with a volume ratio of 3:1 for 10∼20 minutes at room temperature. The distance between the ITO substrate and solution was kept to be at about 2 cm. The patterned ITO was cleaned in flowing water. Then, the cleaning process of the ITO substrate is as follows. First, the patterned ITO substrate was cleaned ultrasonically in chloroform or acetone for 20 minutes at 50°C, after which it was heated to 80°C for 1 hour in a solution made with second distilled deionized water, ammonia, and hydrogen peroxide at a volume ratio of 5:1:1. We sonicated the substrate again in chloroform or acetone for 20 minutes at 50°C. Finally, the substrate was cleaned in isopropyl alcohol and first distilled deionized water for 20 minutes each and dried with nitrogen gas. Thermal vacuum deposition was used in order to deposit the organics at 5×10^{-6} Torr. The deposition was performed using self-made crucible boat stainless steel having excellent thermal conductivity and chemical resistance rather than using conventional thermo stable pyrex boat. After finding the optimum hole size for an evaporation of organics, the Alq_3 was deposited to a thickness of 100 nm at deposition rate of 2.5 Å/s. We measured the electrical characteristics of the device in order to study how surface roughness of Alq_3 layer affects on the efficiency. Al cathode was made by thermal evaporation as well using tungsten boat at 5×10^{-6} Torr. Initial deposition rate of Al was 0.5∼1.0 Å/s up to 10 nm, and then 5 Å/s to 20 nm, and then 15 Å/s all the way up to 100 nm. In addition, the light emitting area was made with the size of $3 \times 5\,mm^2$ using a mask. In order to measure the electrical characteristics of the device, Keithley 2000 multimeter, Keithley 6517 electrometer, and Si-photodiode were used. All devices were measured through a control program of the Lab-view software.

3. Results and Discussion

Figures 1(a) and (b) show the AFM images of surface morphology of Alq_3 layer made by different hole sizes of evaporation boat. Table 1 shows an average roughness of the Alq_3 layer depending on the hole sizes of evaporation boat. Table 1 shows that the average roughness is 1.04 nm for a boat hole size of 1.0 mm. Since this surface roughness is smoother than others, it is expected that the charge injection becomes easier. This may affect on the performance of the device. Figures 2(a) and (b) show the AFM images of surface morphology of Alq_3 thin film made by different



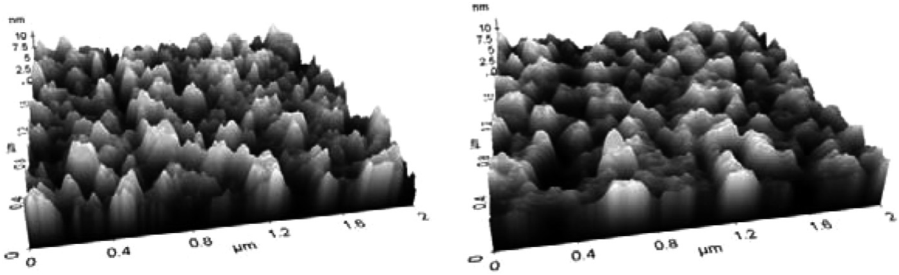
(a) 1.0 mm (b) 3.0 mm

Figure 1. Surface morphology of Alq_3 by several hole sizes of crucible boat.

Table 1. Roughness of Alq_3 by several hole sizes of crucible boat

Hole size of crucible boat [mm]	Average roughness [nm]
0.8	1.20
1.0	1.04
1.5	1.15
3.0	1.31

deposition rates at hole size of crucible boat of 1.0 mm. When the deposition rate is 2.5 Å/s, the average roughness is smoother than others. The average roughness of Alq_3 layer surface is listed in Table 2 for several different deposition rates. When the deposition rate of Alq_3 is 2.5 Å/s, the average roughness is 1.19 and it is smoother than others. This is because a proper evaporative condition helps charge injection from the anode and cathode through a low surface roughness of the organic layers. Thus, charge carriers are well transported to the emission layer which causes an increase in recombination rate. So the light efficiency is obtained. Therefore, by considering the organic light-emitting diodes using an optimal evaporative condition, it is expected that the charge injection from the electrode is easier, and energy barrier



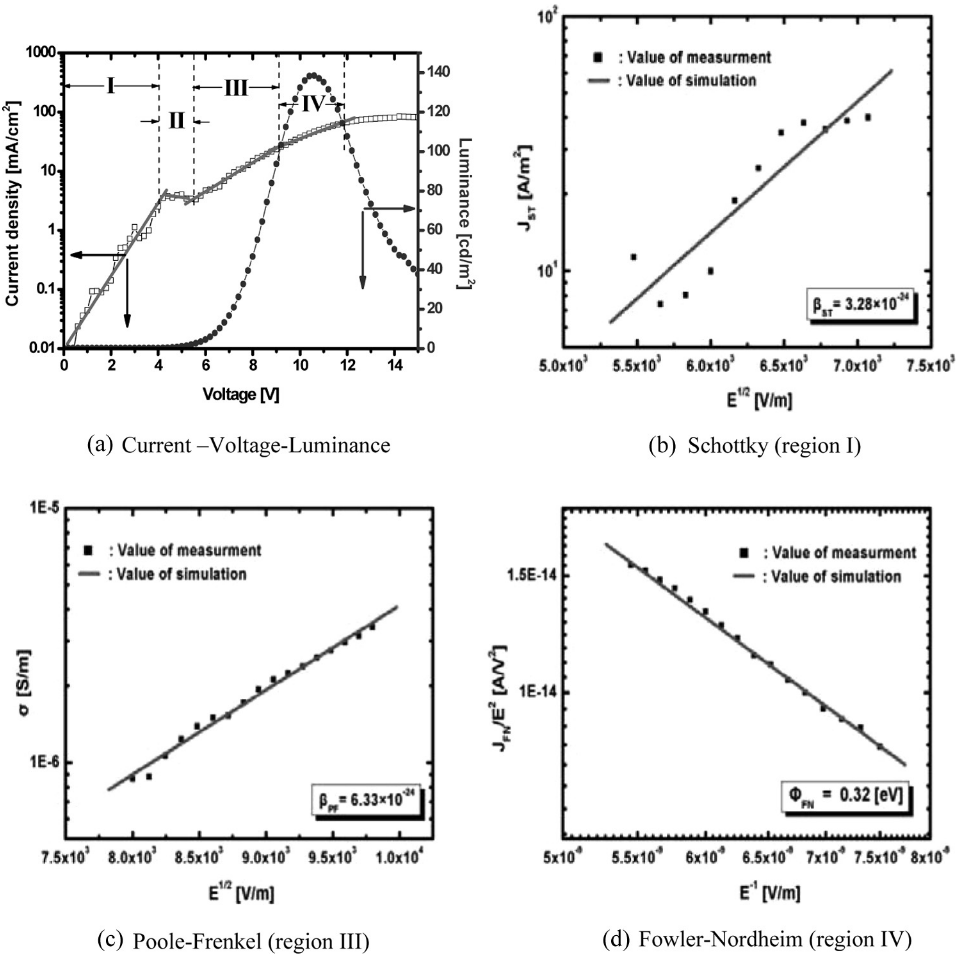
(a) 1.5 Å/s (b) 2.5 Å/s

Figure 2. Surface morphology of Alq_3 by several deposition rates.

Table 2. Roughness of Alq_3 by several deposition rates

Deposition rate [Å/s]	Average roughness [nm]
1.5	1.41
2.0	1.35
2.5	1.19
3.0	1.30

could be lowered. Furthermore, it could contribute to the improvement of the efficiency of the device. Figures 3(a)∼(d) shows the current density and luminance of the device as a function of voltage in a condition of most suitable ITO/Alq_3/Al. We have discovered that the characteristics of conductive current are four different



(a) Current –Voltage-Luminance

(b) Schottky (region I)

(c) Poole-Frenkel (region III)

(d) Fowler-Nordheim (region IV)

Figure 3. Electrical characteristics of ITO/Alq_3/Al made with an optimal deposition condition.

current regions depending on the applied voltage. The conductive current density J_{ST} in region I (Fig. 3(b)), where the electric field is less than 4.0×10^5 V/cm or 4.0 V, can be explained by using Schottky injected conduction. In this region, the current density satisfied the following Eq. (1).

$$J_{ST} = AT^2 \exp\left(\frac{\beta_{ST} E^{1/2} - \phi_D}{kT}\right) \tag{1}$$

Here, β_{ST} is $(e^3/4\pi\epsilon)^{1/2}$. We have obtained that the Schottky coefficient β_{ST} is 3.28×10^{-24} from the slope of straight line of a graph in $\ln J_{ST}$ vs plotted in Figure 3(b). In this region, the current density is proportional to the power of 3.59 of the applied voltage. A region II, where the electric field is less than 4.8×10^5 V/cm is a negative resistance region properties. In this region, the current density is proportional to the power of -1.48 of the applied voltage, specifically, a light emission occurred in this region. A region III (Fig. 3(c)), where the electric field is less than 1.15×10^6 V/cm is a Poole-Frenkel conductive region. In this region, $\ln \sigma$ is proportional to $E^{\frac{1}{2}}$, as shown in Figure 3(c). From a slope of straight line in this figure, we have found that β_{PF} is 6.33×10^{-24} using the Eq. (2). So, we have confirmed that the analysis of conductive mechanism is the following Eq. (2).

$$\sigma = \frac{J_{PF}}{E} = \sigma_0 \exp\left(\frac{\beta_{PF}\sqrt{E}}{2kT}\right) \tag{2}$$

The obtained β_{PF} in Poole-Frenkel region is $1.93\beta_{ST}$, where β_{ST} was obtained in Schottky region. From these analyses, it was confirmed that a fact of $\beta_{PF} = 2\beta_{ST}$ satisfies a theoretical prediction [9,10]. In this region, the current density is proportional to the power of 4.38 of the applied voltage. In order to examine the conductive mechanism in region IV (Fig. 3(d)), tunneling mechanism was applied satisfying the following relation. In order to examine the conductive mechanism in the IV region, the Figure 3(d) shows a relation of $\ln\left(\frac{J_{Fn}}{(E^2)}\right)$ as the 1/E in Schottky region over as conductive region by Fowler-Nordheim using Eq. (3). This region consider that Fowler-Nordheim's conductive current flaws by contribution of electron's tunneling because thickness of barrier is thin as the very high applied electric field of 1.35×10^6 V/cm [11,12].

$$J_{FN} = AE^2 \exp\left(-\frac{\beta}{E}\right) \tag{3}$$

Where, A and B are constants including a work function of the electrode and dielectric and E is the electric field. By applying of Fowler-Nordheim tunneling mechanism, we obtained a barrier ϕ_{FN} of 0.32 eV.

4. Conclusion

The following results were obtained from a research on the electrical conductive mechanism of organic light-emitting diodes depends on the surface roughness of Alq_3 layer. We found that when the hole size of crucible boat and deposition rate of Alq_3 are 1.0 mm and 2.5 Å/s, respectively, an average roughness of the evaporated Alq_3 surface is relatively lower than others. From the analysis of electrical conductive mechanism, it found that there are four different conduction mechanisms. It was found a coefficient β_{ST} in Schottky region is 3.28×10^{-24}, a coefficient β_{PF} in Poole-Frenkel is 6.33×10^{-24}, and a barrier height in Fower-Nordheim is 0.32 eV. And, the obtained β_{PF} is 1.93 β_{ST}, which is in agreement to a theoretical prediction of $\beta_{PF} = 2\beta_{ST}$. A negative resistance region obtained that the current density is proportional to the power of -1.48 of the applied voltage at a field of 4.8×10^5 V/cm. We have confirmed that there are four different conductive mechanisms in the electrical properties of organic light-emitting diodes. Also, the emission of OLED begins at the negative resistance region of current density-voltage, and a maximum luminance shows on the Fower-Nordheim region.

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