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Relation between Preparation Conditions and Characteristics of OLED

Takanori Sasaki, Yoshimine Tanabu, Jin Li, Kaori Suzuki, Katsuyoshi Shinyama, and Shigetaka Fujita

Faculty of Engineering, Department of Electronic Intelligence and Systems, Hachinohe Institute of Technology
88-1, Myo Ohbiraki, Hachinohe, Aomori 031-8501
E-mail:sfujita@hi-tech.ac.jp

In this study on OLEDs, the authors investigated the influence of thickness and surface condition of organic layers on the device characteristics. In this research, we combined deposition rate control and substrate temperature control together to obtain high luminescence, long lifetimes and high efficiency of OLED devices. In addition, a comparison was made of the device characteristics for two different thicknesses of organic layers. These conditions were thicknesses of 15 nm for CuPc, 50 nm for α-NPD and 50 nm for Alq_3 in one case, and 30 nm for CuPc, 100 nm for α-NPD and 100 nm for Alq_3 in the other case. As a result, we succeeded in obtaining high luminescence levels with the deposition rate of 0.1-0.2 nm/s, and the substrate temperature of 48-50 ℃. Moreover, the high brightness at this condition was 1,000 cd/m^2. It is thought that the deposition state was improved with supplementation of molecular alignment energy by combining the deposition rate and the substrate temperature together.

Keywords: OLED, organic electroluminescence, thin film, electrical properties, AFM

1. INTRODUCTION

Recently, OLEDs (Organic Light-emitting Diodes) are being actively researched as the next generation technology for display and lighting devices [1][2]. The research on OLEDs progressed rapidly with the discovery of Alq_3, which can be made into a thin film by vacuum deposition, by Tang et al. in 1987 [3]. An OLED device with Alq_3 as the luminescence layer achieved 1000 cd/m^2 with a driving voltage of less than 10 volts, while conventional devices such as those with anthracene required over 100 volts for emission [4]. Moreover, its lifetime was 100 hours with a starting brightness of 50 cd/m^2, which was longer than that of the conventional devices. Thereafter, the improvement of the device accumulating structure and materials by many researchers has greatly improved the brightness and lifetime.

Under these circumstances, we focused on the preparation process for improvement of device characteristics, and have investigated the influence of thickness and surface condition of organic layers on characteristics [5][6][7]. In our previous study, we succeeded in improving the device characteristics by stabilizing the deposition rate and the substrate temperature [8]. However, the organic-layers were easily burnt off by comparatively low applied voltages, and so a problem remained.

In this research, we investigated the relation among device characteristics, durability, and changing film thickness of organic layers. In addition, we combined deposition rate control and substrate temperature control together to obtain high luminescence, long lifetimes and high efficiency of OLED devices. Moreover, we analyzed the influence of the combination of preparation conditions on device characteristics.

2. DEVICE STRUCTURE OF OLED

In this research, a three-layer structure consisting of a standard two-layer structure with CuPc as a buffer layer to facilitate the injection of positive holes was adopted. The components of the organic layers were a positive hole injection layer (CuPc: Copper Phthalocyanine), a positive hole transport layer (α-NPD: N, N'-Di(1-naphthyl)-N,N'-diphenylbenzidine) and an electron transporting light emission layer (Alq_3: Tris (8-hydroxyquinolinato) aluminum). Molecular structures of the organic materials are shown in Figure 1.

CuPc has heat stability, and it is expected to improve the heat resistance of the device. α-NPD is a thermally stable material, and it hardly crystallizes because it maintains the state of a uniform

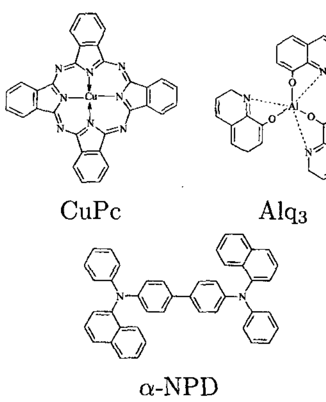


Fig. 1. Organic materials

amorphous film. Alq_3 is also a thermally stable material. An extremely flat film can be made with it and the movement speed of electrons is comparatively high in this material. These organic layers were sandwiched between an ITO (indium-tin oxide) electrode deposited on glass, which acts as the anode, and an aluminum electrode, which acts as the cathode. The structure is shown in Figure 2.

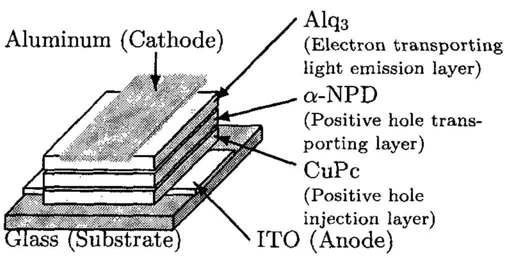


Fig. 2. OLED device structure

This structure is a fairly standard structure. Lifetime and brightness of a device with this structure are lower than recent devices, but preparation is easy and characteristics are comparatively well-known. Accordingly, the influence of process improvement is easy to compare, so we used this structure in this research. Organic materials and aluminum were evaporated using the VPC-1100 High Speed Vacuum Coater (ULVAC KIKO Inc., Ultimate pressure: 5.0×10^{-5}).

The principle of emission is as follows. First, positive holes from the ITO anode and electrons from the aluminum cathode are injected into the organic thin film. Next, carriers move to the counter electrode, and excitons are formed with recombination of electrons and positive holes inside the light emission layer. Finally, light is emitted as the transition energy from the excited state to the ground state of these excitons.

The device prepared in this study has three emission areas, and the size of each area was 12 mm×3 mm.

3. PREPARATION CONDITION

In earlier research, the devices had been formed with a combination of a 15 nm layer of CuPc, a 50 nm layer of α-NPD, and a 50 nm layer of Alq_3. However, this structure had a durability problem. Therefore, the authors doubled the thickness of each layer as a durability improvement in this research, forming a device with a 30 nm layer of CuPc, a 100 nm layer of α-NPD, and a 100 nm layer of Alq_3. Incidentally, the optimum combination for the best ratio of thicknesses of organic layers was shown by Shima [8], and we followed it.

We compared device characteristics under three evaporation conditions with substrate temperature control for 48-52 ℃. The deposition rate for each condition is shown in Table 1.

Table 1. Preparation conditions of organic layer

	Condition 1	Condition 2	Condition 3
Thickness of Layer [nm]	CuPc : 30, α-NPD : 100, Alq_3 : 100		
Substrate Temp. [℃]	48-50		
Deposition rate [nm/s]	0.01-0.03	0.05-0.15	0.15-0.30

Additionally, we compared device characteristics and surface conditions by control precision of substrate temperature at low-precision (±5 ℃) and high-precision (±1 ℃).

4. RESULTS AND DISCUSSION

Measurement results under three evaporation conditions are shown in Figures 3 and 4.

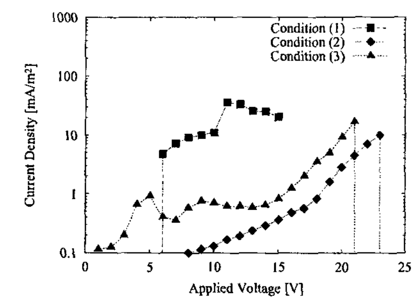


Fig. 3. Voltage-Current density characteristics for three deposition rate conditions

As shown in Figure 4, the highest brightness for Condition 1 was 36.14 cd/m^2 as 11 volts were applied. Compared with Condition 1, the highest brightness values for Conditions 2 and 3 were much higher at about 1000 cd/m^2 each. In addition, when the voltage and current density which

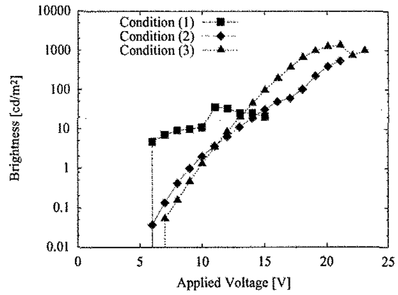


Fig. 4. Voltage-Brightness characteristics for three deposition rate conditions

gave the same brightness for Conditions 2 and 3 were compared as shown in Figure 3, hardly any difference was found in the luminous efficacy. Luminous efficacy is defined by the following equation.

$$\eta = 4\pi \frac{B}{E \cdot D_I} \qquad [\mathrm{lm/W}] \qquad (1)$$

B: Brightness $[\mathrm{cd/m^2}]$
E: Applied Voltage [V]
D_I: Current Density $[\mathrm{A/m^2}]$

For example, luminous efficacy is 24.5 lm/W at 18 V for Condition 2 and 21.8 lm/W at 15 V for Condition 3, and these are higher than for Condition 1 (highest efficacy is 0.5 lm/W). The highest brightness shown was 1341 $\mathrm{cd/m^2}$ for Condition 3, an improvement over the former device.

When these results and current findings are considered together, it has been understood that there is a suitable combination of substrate temperature and deposition rate in evaporation. In this case, Condition 3 (0.15-0.3 nm/s) was most suitable. Incidentally, device lifetime is under measurement in detail. The former device had burnt off after emitting light for about two hours, and it was hardly measured enough. But the device which was evaporated under the new preparation conditions maintained sufficient brightness for 24 hours or more, and adequate measurement became possible.

Next, the results of characteristics measurement for low-precision and high-precision substrate temperature control were compared as shown in Figures 5 and 6. The emission conditions for these are shown in Figures 7 and 8. There was a one order of magnitude difference in brightness with high-precision control compared with low-precision control when 15 volts were applied as shown in Figure 6. It appears clearly in Figures 7 and 8.

When emission conditions are observed in Figures 7 and 8, non-uniformity of emission is conspicuous at 9-13 V for low-precision control. Figure 9 shows the result of measurement of surface

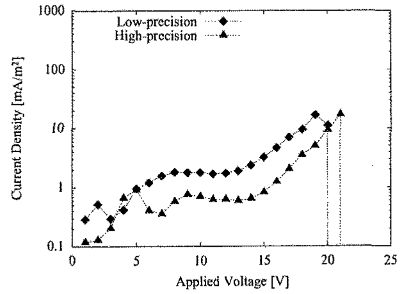


Fig. 5. Voltage-Current density characteristics for two substrate temperature control precision conditions

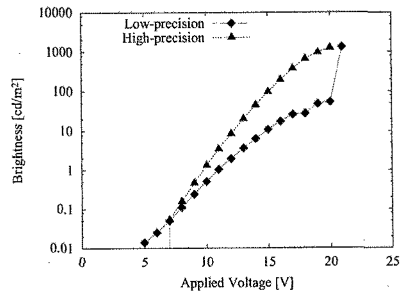
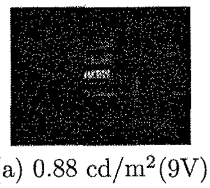
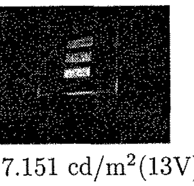
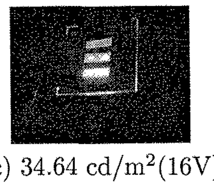


Fig. 6. Voltage-Brightness characteristics for two substrate temperature control precision conditions


(a) 0.88 $\mathrm{cd/m^2}$(9V)


(b) 7.151 $\mathrm{cd/m^2}$(13V)


(c) 34.64 $\mathrm{cd/m^2}$(16V)

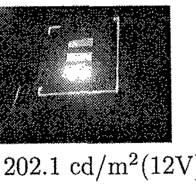
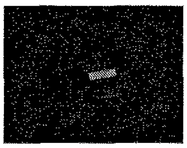
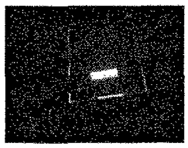

(d) 202.1 $\mathrm{cd/m^2}$(12V)

Fig. 7. Emitting condition of low-precision temprature control

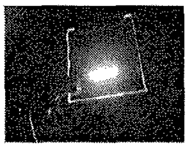
condition using AFM with consideration of this non-uniformity. The average of surface roughness and maximum height difference is shown in Table 2. The surface condition of the device for the low-precision control is inhomogeneous as shown in Table 2. In particular, the height difference is very large at about 800 Å, and there are over 100 molecular layers when the molecular size of Alq_3 is considered. It is thought that the non-uniformity of emission in Figure 7-(a), (b) originates from this surface condition. On the other hand, it is thought that the organic layer is evap-



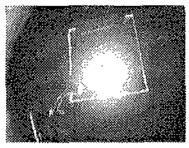
(a) 0.462 cd/m^2(9V)



(b) 20.03 cd/m^2(13V)

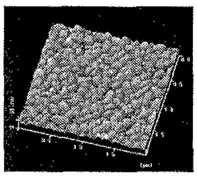


(c) 191.4 cd/m^2(16V)

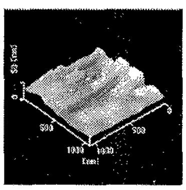


(d) 1341 cd/m^2(21V)

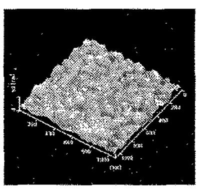
Fig. 8. Emitting condition of high-precision temperature control



(a) ITO electrode



(b) Organic Layers (low-precision temperature control)



(c) Organic Layers (high-precision temperature control)

Fig. 9. Surface condition (AFM image)

Table 2. Difference of surface condition

	ITO	Organic Layers	
		low precision	high precision
Average of surface roughness[nm]	5.015	6.514	2.456
Maximum height difference[nm]	20.92	79.92	14.31

orated uniformly because the order of roughness and height difference of the high-precision controlled device is similar to that of ITO.

From these results, it has been understood that the influence on device condition of surface temperature is very important and it is necessary to have sufficient precision of temperature control. Additionally, it is important to decrease the roughness of the ITO electrode for smoothing of the deposition surface, and for that purpose, it is necessary to improve the etching method and substrate washing process to adequately remove dust and scratches.

5. CONCLUSION

We examined the influence of deposition rate and substrate temperature control precision on the device characteristics of OLEDs. Because the level of vacuum was not high enough, the brightness of the device in this research is not brighter than that of other similar devices, but the effect on device characteristics of different preparation conditions was confirmed. Therefore, it was understood that appropriate control of the substrate temperature and deposition rate is necessary to make a device with high characteristics. A reason for this is thought to be that the energy to diffuse and to deposit evaporated organic materials uniformly on a substrate is supplemented by evaporation under suitable conditions.

For the control precision of substrate temperature, device characteristics and emission state were influenced by the change of surface conditions of organic layers due to control precision. It has been found that organic layers can be deposited almost uniformly with sufficient precision control. The improvement of the method of washing the substrate and suitable preparation conditions are needed for further smoothing of the layer surface.

Hereafter, we will examine the device state by analyzing the surface composition using XPS and crystalline level using XRD, and analyze the deterioration process.

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