Development of Phosphorescent White OLED with High Power Efficiency and Long Lifetime

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Abstract

White OLED device with extremely high power efficiency and long lifetime was developed in which blue, yellow-green and red phosphorescent emitters were used. The performances achieved were 64 lm/W, and 10000 hours of lifetime at initial luminance of 1000cd/m^2 with a light out-coupling technique. The device also exhibited the good durability important in practical use. New technologies, such as blue phosphorescent materials and a sophisticated organic layer structure were applied to the device. Our hope is that these technologies will open the door to the practical use of OLEDs as lighting sources.

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1 Introduction

Because of recent environmental protection and energy supply situations, eco-friendly lighting sources of low energy consumption are in demand. An enormous amount of research effort goes into the field, and, over the past several years, particular attention has been paid to inorganic and organic light-emitting diodes as new lighting source candidates.

Organic light-emitting diodes (OLEDs) are regarded as a powerful candidate because they are an area lighting source and can be driven at voltages as low as just several volts, and, further, they contain no materials (e.g. mercury) that are harmful to the human body or the environment. As a lighting source for illumination or display backlight, a white light is usually required. It is well known that the use of plural light emissive materials such as blue, green, and red is useful in obtaining white emission.

This raises several questions: When a white OLED is used as lighting sources, how much efficiency is required? How much external emission quantum efficiency (EQE) and drive voltage are needed? It seems that a power efficiency nearly matching that of the current lighting sources is necessary to replace them with OLED lighting, depending on the application of the light source. The power efficiency of current lighting sources in general is about 10 to 20lm/W for electric bulbs and 60 to 90lm/W for fluorescent lamps.

Fig.1 shows the relationship between power efficiency and drive voltage at given EQE values for an OLED which emits warm white color. From this figure, as an example, it is understood that a drive voltage of 4V and an EQE of 35% are necessary to obtain the power efficiency of 60lm/W. Because the lowest drive voltage of a white OLED is limited to about 3V by triplet excitation energy of the blue phosphorescent material, an EQE of over 25% is needed to obtain at least 60lm/W.

OLEDs fall roughly into two classifications defined

by their emission mechanism: fluorescent OLEDs and phosphorescent OLEDs. It is well known that the phosphorescent OLED is advantageous from the aspect of emission quantum efficiency [1]. The emission quantum efficiency of fluorescent OLED is only one quarter that of a phosphorescent OLED, and, theoretically, a phosphorescent OLED can reach 100%.

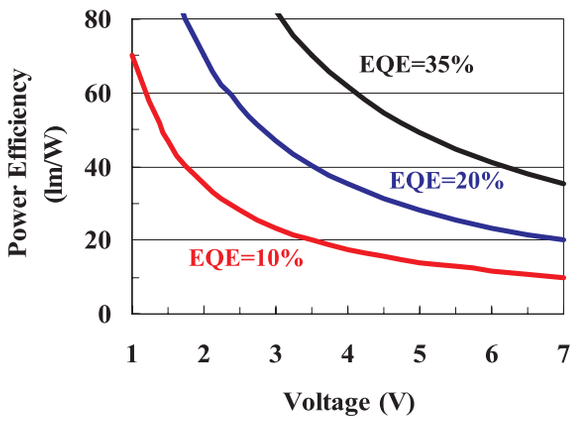

Fig.1 Power efficiency vs. driving voltage

Therefore, developing a phosphorescent OLED seemed the better course in obtaining higher power efficiency. Indeed, Y. Tung et al. reported a phosphorescent white OLED with high efficiency [2]. Its EQE reached 16%, and the power efficiency was 18.4lm/W. However, there was a severe problem in obtaining a phosphorescent white OLED: the lifetimes of blue phosphorescent materials have been too short for practical application in a phosphorescent OLED.

Recently, however, an improvement of the lifetime of blue phosphorescent materials was reported. M. S. Weaver et al. reported blue phosphorescent OLEDs with improved operating stability [3]. B. W. D'Andrade et al. reported a phosphorescent white OLED with 14% EQE and a long lifetime [4]. However there is still estrangement with theoretical maximum EQE of a phosphorescent OLED.

We engaged in the development of blue phosphorescent materials with a long lifetime, and, at the same time, we pushed forward with development of an organic layer structure to obtain higher power efficiency. As a result, we were able to develop a phosphorescent white OLED of high performance. **Fig.2** shows an example of the white OLED device we developed. In this paper, we discuss the major performances of and an outline of the technology incorporated in our white OLED device.

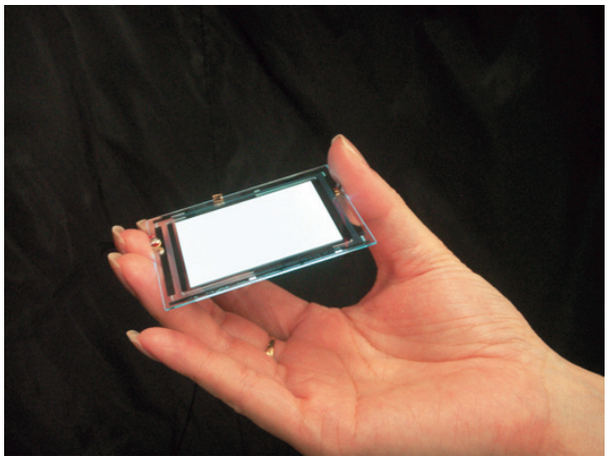

Fig. 2 Photograph of the white OLED device developed

2 Device structure and evaluation

The structure of our phosphorescent white OLED device is shown schematically in **Fig.3**. Organic layers were evaporated on an indium tin oxide (ITO) anode whose thickness was 110nm under pressure of less than 4×10^{-5} Pa. A hole injection layer (HIL), hole transport layer (HTL), emissive layers, inter layer, and electron transport layer (ETL) were evaporated on the anode in that order. An aluminum (Al) cathode with a thickness of 100nm was evaporated on the electron transport layer.

Emissive layers have blue, yellow-green and red phosphorescent dopants and host material. The blue dopant and the host material were developed and synthesized by our laboratory. The yellow-green (YD-85) and red (RD-61) phosphorescent dopants were obtained from Universal Display Corporation. The electron transport material used in the ETL was also developed and synthesized by our laboratory.

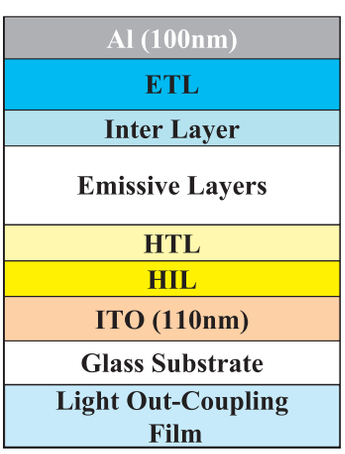

Fig. 3 Device structure of the white OLED

Glass cap encapsulation was accomplished using UV hardening epoxy glue in a pure, dry nitrogen atmosphere. Oxygen concentration was less than 1 ppm, and vapor concentration was less than 0.1ppm in the encapsulation environment. An absorbent was included in the device.

To improve the light out-coupling efficiency of the device, a diffusive light out-coupling film was affixed on surface of the glass substrate on the side of the device opposite the organic layers.

Emission areas of our devices were 4mm^2 or 15cm^2. The efficiencies were nearly the same in both devices. Lifetime was measured for 4mm^2 device.

The current-voltage-luminance characteristics and emission spectra of the devices were measured by spectrophotometer (CS1000: Konica Minolta Sensing, Inc.). Light distribution was measured to calculate power efficiency. Device lifetime was defined as the time taken to reduce luminance to half of the initial luminance under continuous drive conditions. Storage stability was evaluated via an accelerated test condition of 85℃.

3 The white OLED's performance

3．1 Major characteristics

Major characteristics and performance of our white OLED are shown in Table 1. Our white color device had a color temperature of 4500K, CIE1931 color coordinates of (0.37, 0.42), and an EQE of 34%, A drive voltage of 3.6V and a power efficiency of 64lm/W were obtained at a luminance of 1000cd/m^2. The light distribution was almost Lambertian. The lifetime of the device was estimated to be 10000 hours at an initial luminance of 1000cd/m^2. These performances were virtually unchanged after over 500 hours of storage at 85℃.

Table 1 Performance of our phosphorescent white OLED at 1,000cd/m^2

Characteristics at 1000cd/m^2	Performance
Power efficiency (lm/W)	64
EQE (%)	34
Drive voltage (V)	3.6
Lifetime $T_{L50\%}$ (h)	10000
Color coordinates (x, y)	0.37, 0.42
Storage stability at 85℃ (h)	>500

3．2 Efficiencies and lifetime

Fig.4 shows the dependence of EQE on luminance. It shows that an EQE of 34% was obtained at 1000cd/m^2. Though the EQE falls slightly on the high luminance side of the peak at approximately 100cd/m^2, the dependence of the EQE on the luminance is small. Therefore, we can assume that the organic layer structure maintains good carrier balance over a wide range of luminance.

The data shown in Fig.4 relates to the device with light out-coupling film shown in Fig.3. By affixing the light out-coupling film, the amounts of light emitted outside of the device rose about 1.6 times. The EQE of the device without the light out-coupling film was 20% at 1000cd/m^2.

Fig.5 shows the dependence of luminance on drive voltage. As seen in the figure, a drive voltage of about 3.6V at 1000cd/m^2 was obtained.

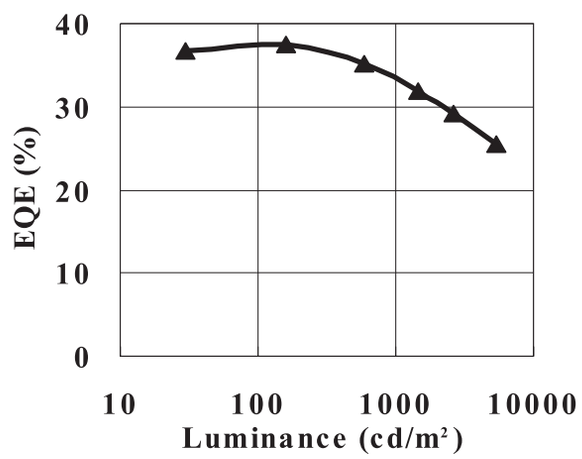

Fig. 4 Dependence of EQE on luminance of the device shown in Fig. 3.

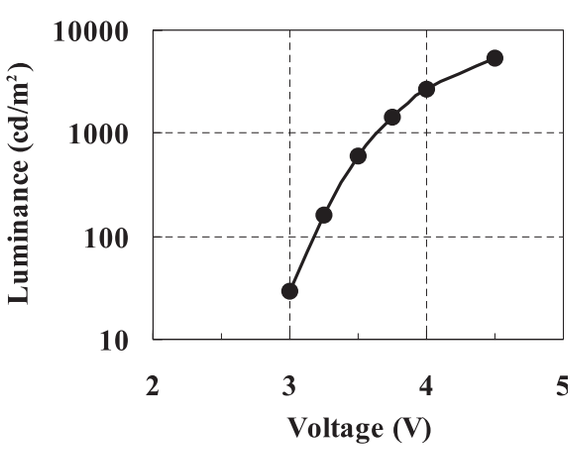

Fig. 5 Dependence of luminance on voltage of the device shown in Fig. 3.

Fig.6 shows the dependence of power efficiency on luminance. By achieving the high EQE and the low drive voltage shown in Fig.4 and Fig.5, a power efficiency of 64lm/W was obtained at 1000cd/m^2. Power efficiency reached about 90lm/W at 100cd/m^2.

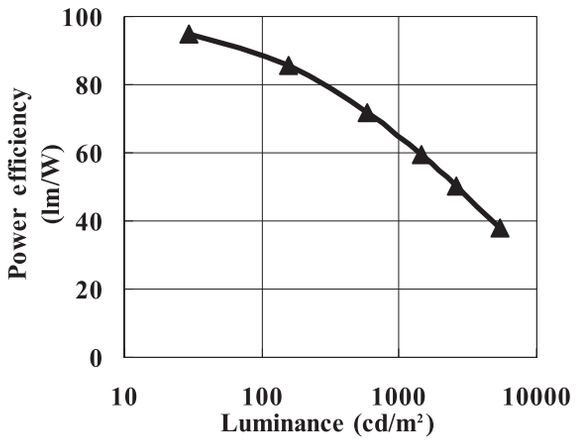

Fig. 6 Dependence of power efficiency on luminance of the device shown in Fig. 3.

Fig.7 shows the operating stability of the device shown in Fig.3 under continuous driving conditions. The initial luminance was 4800cd/m^2. It is known that the relationship between lifetime and initial luminance is expressed by the following equation (1).

$$t(L_1) = t(L_2) \times (L_2 / L_1)^X \quad (1)$$

Here, $t(L_1)$ and $t(L_2)$ are lifetimes measured at initial luminance L_1 and L_2 respectively. Here x is an acceleration coefficient, and its value is generally between 1.5 and 2. Assuming that x is 1.5, which is a conservative estimate, a lifetime of 1000 hours measured with an initial luminance of 4800cd/m^2 is equivalent to the lifetime of 10000 hours with an initial luminance of 1000cd/m^2.

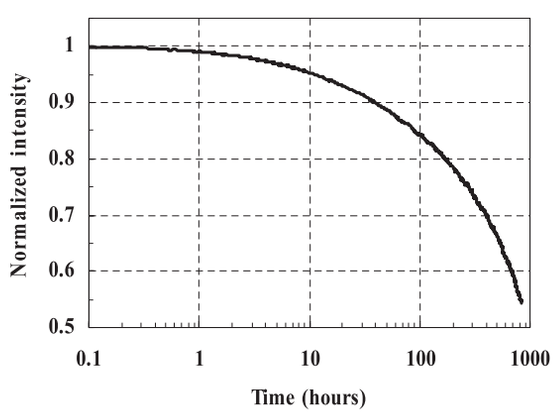

Fig. 7 Operational stability under continuous driving conditions. Initial luminance was 4,800cd/m^2 of the device shown in Fig.3.

3．3 Storage stability

Storage stability is also important in practical use. Fig.8 shows the storage stability of luminance and color measured under an accelerated test condition of 85℃. Luminance and color were measured at a constant current of 2.5mA/cm^2 and at room temperature. In this figure, the dots indicate luminance and the triangles indicate color shift ⊿E, where ⊿E as defined by equation (2) indicates the degree of color change.

$$\Delta E = \sqrt{(x(t)-x(0))^2+(y(t)-y(0))^2} \quad (2)$$

Here, x(t) and y(t) indicate color coordinates x and y in a CIE1931 color diagram at storage time t, while x(0) and y(0) indicate color coordinates x and y before storage.

The color shift ⊿E was less than 0.005 after storage for 520 hours at 85℃, and the change in luminance was less than 1%.

In addition, no outbreaks of dark spot or decreases in the emission area were observed, as shown in the photographs in Fig.9. In Fig.9, on the left our device is in a driving state before storage, while in the right the device in a driving state after storage of 520 hours at 85℃.

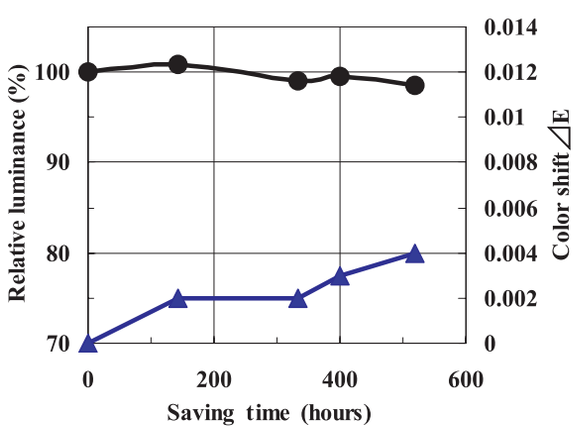

Fig. 8 Storage stability of luminance and color at 85° C. Dots (●) indicate luminance and triangles (▲) indicate color shift ⊿E.

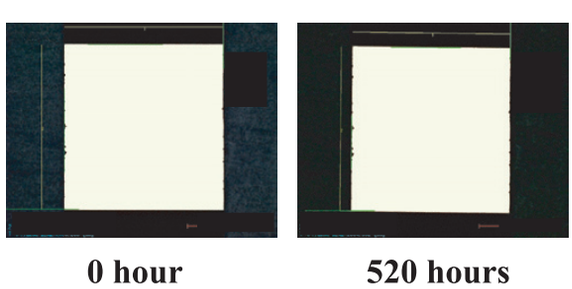

Fig. 9 Photographs of OLED in driving state before and after durability test

4 Key technologies

4．1 Material

The most important materials technology involved with our device are blue phosphorescent materials. Table 2 shows the performances of FIr(pic), which is a typical blue phosphorescent dopant, and KMBD-342, a blue phosphorescent dopant developed by our laboratory. The data shown in Table 2 were obtained for a blue OLED device whose layer structure was ITO (110nm) / HIL / α-NPD / blue emissive layer / BAlq / LiF / Al (100nm).

Table 2 Performance of blue phosphorescent materials

Material	Emission peak wavelength (nm)	EQE (%)	Current efficiency (cd/A)	Lifetime (hours @300cd/m²)
FIr(pic)	469	16	31	<100
KMBD-342	472	17	42	16000

Even though the emission efficiency of FIr(pic) is high, its lifetime is well known to be too short for that material to be applied to practical devices.

We found a dopant, KMBD-342, and a host material which was suitable for KMBD-342. As shown in Table 2, KMBD-342's EQE was 17%. Although KMBD-342's EQE was not much higher than that of FIr(pic), the lifetime of KMBD-342 was dramatically higher than that of FIr(pic), reaching a 16000-hour lifetime with an initial luminance of 300cd/m². These blue dopant and host materials contributed greatly to obtaining the lifetime of 10000-hour lifetime of our white OLED.

4．2 Organic layer design

To obtain high power efficiency, it is necessary to simultaneously raise EQE and lower drive voltage. As well as material technology described above, the layer design of the white emissive layers contributed to raising EQE, and the layer design of the ETL contributed greatly to lowering the drive voltage.

4．2．1 White emissive layers

We designed the emissive layers to achieve 20% EQE without a light out-coupling technique. The individual EQEs of the blue, yellow-green, and red dopants in a layer structure which was similar to the white OLED were 17%, 17% and 12% respectively. If each dopant in the white OLED emits light with the above EQE, the EQE of the white light is calculated to be only about 15%, well below 20%, which is generally said to be theoretical maximum EQE of phosphorescent OLED.

However, we found that we could raise EQE by optimizing the carrier trapping property and using an energy transfer mechanism in the emissive layer, including the red dopant, which had the lowest EQE among the dopants that we used. By using such a techique, as a result, the EQE of our white OLED reached 20% without light out-coupling film even though EQE of each individual dopant was less than 20%. Further, the EQE of our white OLED reached 34 % when we utilized a light out-coupling film as shown in Fig.4.

4．2．2 Carrier transport layers

Doping technology was introduced to the electron transport layer to achieve low drive voltage, i.e. our electron transport layer was so-called n-type electron transport layer. The drive voltage fell to about 3V at 1000cd/m² as shown in Fig.10. This contributed greatly to the improvement of power efficiency.

In Fig.10, the dots indicate the luminance-voltage (L-V) characteristics of the device, which uses the developed carrier transport materials, and the squares indicate L-V characteristics of a device using conventional carrier transport materials, such as CuPc as hole injection material, α-NPD as the hole transport material, Alq_3 as the electron transport material and LiF as the electron injection material. Both devices were green phosphorescent OLEDs using GD48 as a green phosphorescent emitter and CBP as the host material

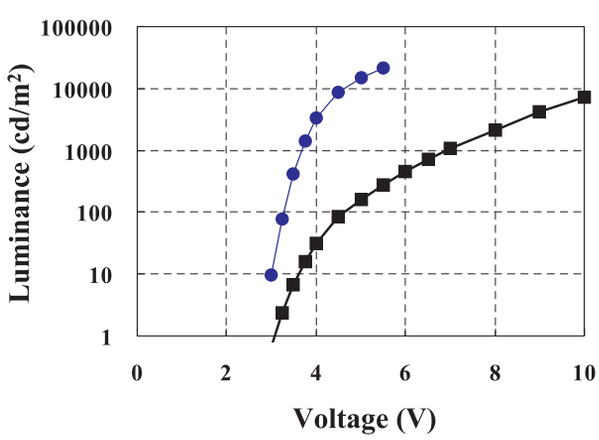

Fig. 10 Dependence of luminance on voltage

in the emissive layer. GD48 was obtained from Universal Display Corporation.

5 Future works

As shown in **Fig.11**, the performance of our OLED device surpassed that of electric bulbs and reached the domain of fluorescent lamps. Although further study

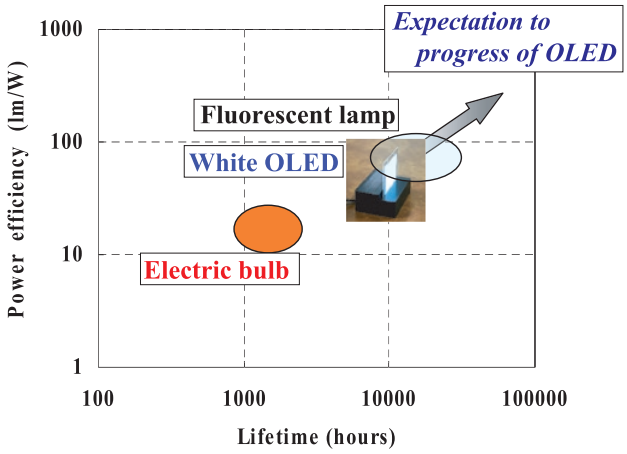

Fig. 11 Light source trends: combined power efficiency and lifetime

will be needed in the future, encouraging characteristics of durability, such as the storage stability essential to practical use, were obtained.

In the future, further improvement of power efficiency and lifetime will be necessary if higher luminance is to be achieved. But gaining higher luminance will establish a foothold in the proliferation of this technology, and we hope that the technologies discussed in this paper lead to the practical use of white OLEDs as lighting sources for illumination, backlights, and more.

6 Conclusion

We developed a phosphorescent white OLED device with a power efficiency of 64lm/W and a lifetime of 10000 hours at an initial luminance of 1000cd/m^2. An EQE of 34 % and a low drive voltage of near 3.6V at 1000cd/m^2 were realized. This performance was due to the development of organic materials, such as blue phosphorescent and electron transport materials, and to progress in organic layer structure design.

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(An additional note ; The contents described in this paper was presented at Society for Information Display 2007 International Symposium (Digest of technical papers Vol.38, p.1018))