Development of Long Lifetime and High Performance Organic Light Emitting Diode Display with Wide Temperature Range

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A long lifetime organic light emitting diode (OLED) display at high temperature was developed with optimization of each layer organic materials. We clarified that charge-carrier balance between hole and electron inside the emission layer (EML) affects the lifetime of OLEDs, especially for blue-OLED. We also clarified that the lifetime becomes long with increasing the density of hole in the EML. The lifetimes of the blue-, green, and red-OLEDs developed in this study were achieved over 1,000 hours at 85 °C under a driving of 600 cd/m² white luminance. A prototype 12.3 inch flexible OLED display was fabricated with using the new electron-blocking layer, hole-blocking layer, and hole-transport layer materials. We believe that the OLED display that we developed would be useful for automotive application.

Keywords: Organic light emitting diode display, Lifetime, High temperature driving, Automotive application

1. Introduction
Organic light emitting diode (OLED) displays show great display performances such as high contrast ratio, wide color gamut, wide viewing angle, and etc. [1-4]. Furthermore, flexible, bendable, and rollable OLED displays are being actively developed in recent years [5-8]. Hence the market of the OLED display has been rapidly growing for many applications such as smart phones, tablets, televisions, and head-mounted displays. In addition, a demand for displays toward automotive application has also become large, and most of the automotive displays used recently are liquid crystal displays (LCDs). The LCDs, however, show low contrast ratio, slow response speed, and narrow viewing angle [9]. Moreover, the stability of the LC material at both high and low temperature is low in principle because nematic phase of the LC material is shifted to other phases. Therefore, the LCDs for the automotive application are limited. On the other hand, the stability of organic materials for the OLED is relatively stable in wide temperature range since phase transition does not take place in the temperature range between -40 °C and 100 °C. Thus, the OLED displays are potentially suitable for the automotive application from the standpoint of not only performances of OLED device (OLED) but also the stability of the organic materials against environmental temperature. However, it is known that lifetime of OLED at high temperature is relatively short [10-12]. Hence it is necessary to improve the lifetime in order to go into the market of the automotive field.

On the basis of the above background, we attempted to improve the lifetime of OLED, especially blue (B)-OLED, by optimizing the organic materials. A device structure of the OLED used in this study is depicted in Fig. 1. For improvement of the lifetime, charge-carrier balance between hole and electron carriers in an emission layer (EML) was optimized. It is generally known that a hole-dominant OLED, especially B-OLED, shows relatively long lifetime compared with an
electron-dominant OLED [13,14]. Thus, we firstly optimized organic materials for electron-blocking and hole-blocking layers (EBL and HBL) with the B-OLED, which are adjacent to the EML. Then, we optimized a material for a hole-transport layer (HTL) in order to further improve the lifetime. And finally, we also attempted to evaluate the lifetimes of green- (G-) and red- (R-) OLEDs.

Fig. 1. Device structure of the OLED used in this study.

2. Experimental
2.1. Fabrication of OLED devices (OLEDs)
The B-, G-, and R-OLEDs were each fabricated for evaluation of lifetimes. Six organic layers produced from organic materials were prepared with vacuum deposition technique under appropriate temperatures. The reflective anode and cathode layers were produced from indium-tin oxide and Mg/Ag, respectively. In order to evaluate a hole transport property of the HTL materials, hole-transport devices (HTDs) were also fabricated in a manner similar to the fabrication of the OLEDs. The structure of the HTD is anode/HIL/HTL/EBL/cathode.

2.2. Organic materials
We used the vacuum deposition type organic materials in this study. Highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO) levels of each organic materials used for B-OLED are shown in Fig. 2.

2.3. Measurements
The lifetimes of the OLEDs were measured under 50 mA/cm² for the B- and R-OLEDs, and 30 mA/cm² for the G-OLED at 25 °C, 70 °C, and 85 °C. Lifetime measurements at 25 °C were carried out with an EAS-10G, and those at 70 °C and 85 °C were carried out with an EAS-61F developed by System Engineers’ Co.

Fig. 2. HOMO and LUMO levels of the organic materials for the B-OLED used in this study.

3. Results and discussion
3.1. EBL and HBL materials for B-OLED
The EBL and HBL materials were firstly optimized with using the B-OLED. We selected two EBL materials (the EBL-1 and EBL-2) and three HBL materials (the HBL-1, HBL-2, and HBL-3) with different HOMO and LUMO levels, as shown in Fig 2. Figure 3 shows decay profiles of relative luminance measured at 70 °C for the B-OLEDs with using the EBL-1 and EBL-2, respectively. Other organic layers were fixed at the HTL-1 and the HBL-1. The lifetime of the B-OLED with the EBL-2 shows twice longer than that of the B-OLED with the EBL-1. The HOMO level of the EBL-2 is slightly deeper than that of the EBL-1, suggesting that a hole-injection into the EML is relatively easier for the EBL-2 compared with the EBL-1.

Then, a dependence for the lifetime of the HBL material was evaluated in the same experimental condition. The material EBL-2 was used as the EBL material. The decay profiles of the B-OLEDs...
measured at 70 °C are shown in Fig. 4. The result indicates that the lifetime was further improved with using the HBL-2 and HBL-3. In particular, the lifetime of the B-OLED with the HBL-3 became almost twice long compared with that of the B-OLED carrying the HBL-1. The result suggests that the lifetime of the B-OLED becomes long as the LUMO level of the HBL material becomes deep. When the LUMO level is deeper than that of the EML, the rate constant of an electron-injection into the EML would be relatively small [15]. Therefore, one can estimate that the density of the free-electron in the EML would be low, leading to a long lifetime since the density of the free-electron is comparably low. In contrast, the rate constant of the electron-injection into the EML becomes high when the LUMO level of the HBL is shallower than that of the EML. In this case, there is a possibility that the density of the free-electron would be high in the EML owing to the injection of a large amount of electron, leading to a comparably short lifetime [12]. Figure 5 presents a schematic diagram for a mechanism of hole and electron injections into the EML. The lifetime of our B-OLED becomes long with relatively increasing the ratio of the hole compared with the electron in the EML. This is qualitatively coincident with the result reported by Böhmer et al. [13,14].

3.2. HTL material

As described above, the lifetime of the B-OLED becomes long for the hole-dominant OLED compared with the electron-dominant OLED [13,14]. The fact gives us estimation that the hole-transport property of the HTL also affects the lifetime of the OLED. We assumed that the lifetime becomes long with increasing the hole-transport property of the HTL. Therefore, hole-transport property of two HTL materials (the HTL-1 and HTL-2) were firstly attempted to evaluate with using the HTDs though these two materials show almost same HOMO and LUMO levels (see Fig. 2). Figure 6 shows the $J$-$V$ curves for the HTDs with using two HTL materials. The current density of the HTD carrying the HTL-2 shows larger than that carrying the HTL-1, implying that the hole-transport property of the HTL-2 is larger than that of the HTL-1. Hence, there is a possibility that the lifetime would be further improved by using the HTL-2 instead of the HTL-1 because the amount of hole in the EML is estimated to be increased by using the HTL-2.
Figure 7 shows the decay profiles of relative luminance measured at 70 °C for the B-OLEDs with using the HTL-1 and HTL-2, respectively. Other organic layers were fixed at the EBL-2 and the HBL-3. With using the HTL-2, further improvement of the lifetime was obtained. Thus, we confirmed that the charge-carrier balance of the OLED strongly affects the lifetime, and the lifetime becomes long as the OLED becomes the hole-dominant.

Fig. 7. Decay profiles of the B-OLEDs with the HTL-1 and HTL-2 measured at 70 °C.

3.3. Lifetimes for B-, G-, and R-OLEDs at 85 °C

With using the new selected EBL, HBL, and HTL materials, the G- and R-OLEDs were also fabricated. In the case of the HBL and the HTL, same materials with the B-OLED (the HBL-3 and the HTL-2) were used because the HBL and the HTL are both the common layers among the B-, G-, and R-OLEDs. In contrast, the materials for the EBL were severally optimized for the G- and R-OLEDs in a manner similar to the case of the B-OLED. The characteristics of each color OLEDs evaluated at 25 °C are listed in Table 1. The lifetimes at 85 °C were then evaluated because the temperature inside the cars becomes extremely high in a summer season. Figure 8 indicates the decay profiles of the B-, G-, and R-OLEDs. The result indicates that the significant improvement of the lifetimes was obtained with using the new EBL, HBL, and HTL materials. In particular, the improvement for the lifetime of the B-OLED is prominent, anticipating that the lifetime of the B-OLED is remarkably affected by the charge-carrier balance between the hole and electron.

Table 1. Characteristics of the B-, G-, and R-OLEDs under 10 mA/cm² at 25 °C.

| OLEDs | Blue | Green | Red |
|-------|------|-------|-----|
| cd/A  | 5.2  | 130   | 40  |
| Chromaticity (x, y) | (0.14, 0.05) | (0.24, 0.72) | (0.78, 0.31) |

Fig. 8. Decay profiles of the (a) B-, (b) G-, and (c) R-OLEDs measured at 85 °C.

3.4. Estimation of lifetime for the automotive display

For the automotive display application, enough level of the lifetime at high temperature such as 85 °C is crucially important. Therefore, we estimated the lifetime at 85 °C under a real use as the automotive display. The lifetime under the real use is defined in the following Eq. (1) [16],

$$\text{Lifetime}_{\text{real}} = \text{Lifetime}_{X} \left( \frac{X}{A_{\text{real}}} \right)^k \quad (X = 50 \text{ or } 30) \quad (1)$$

where $\text{Lifetime}_{\text{real}}$ and $\text{Lifetime}_{X}$ indicate the lifetimes under current densities (mA/cm²) of the real use $A_{\text{real}}$ and $X$, respectively. $A_{\text{real}}$ indicates the current density of the real use, and $k$ indicates an accelerating factor. We attempted to estimate the $\text{Lifetime}_{\text{real}}$ for the B-, G-, and R-OLEDs with using the above relation. As the real use, 600 cd/m² luminance of white color with the chromaticity (0.31, 0.32) was selected, and the values of $A_{\text{real}}$ for the B-, G-, and R-OLEDs were estimated in the case of using the OLED panel that we developed; details of the OLED panel specifications developed by us are described below. In addition, the $k$ value of the B-OLED was determined from the above equation by using the lifetimes under the driving conditions.
of 30, 50, and 100 mA/cm², as shown in Fig. 9. We obtained the $k$ value for the B-OLED at 85 °C is 1.31. The $k$ values for the G- and R-OLEDs were also determined in a manner similar to this method.

![Decay profiles of the B-OLED with the EBL-2, HBL-3, and HTL-2 materials under driving conditions of 30, 50, and 100 mA/cm² measured at 85 °C.](image)

We defined the $\text{Lifetime}_{\text{real}}$ as the time for the luminance to decay to 80% of its initial value. The obtained lifetimes for the B-, G-, and R-OLEDs are summarized in Table 2. The result indicates that the values of $\text{Lifetime}_{\text{real}}$ under the 600 cd/m² white luminance show over 1,000 hours. Thus, we confirm that the OLEDs developed in this study show enough level of the lifetime for the automotive display.

![Photographs of the 12.3 inch flexible OLED displays, (a) bendable type, and (b) “S”-character type.](image)

3.5. Fabrication of OLED Display

With using the new selected OLED materials, we fabricated a prototype 12.3 inch flexible OLED display, as shown in Fig. 10. The prototype OLED display showed good brightness uniformity. The specifications of the prototype OLED display are listed in Table 3. The fabricated OLED display could be used in a wide temperature range from -40 °C to 95 °C. In addition, since a polyimide substrate was used, the OLED display shows high flexibility, as shown in Fig. 10. Finally, we expect that the developed OLED display is useful for the automotive application.

### Table 2. Estimated lifetimes of the B-, G-, and R-OLEDs with white luminance of 600 cd/m² at 85 °C.

| TE-OLED | Current density (mA/cm²) | Lifetime (h) | Current density for 600 cd/m² white (mA/cm²) | $k$ | Lifetime for 600 cd/m² white (h) |
|---------|---------------------|-------------|------------------------------------------|-----|---------------------------------|
| Blue    | 50                  | 190         | 6.6                                      | 1.31| 2.7x10³                         |
| Green   | 30                  | 170         | 6.3                                      | 1.42| 1.5x10³                         |
| Red     | 50                  | 230         | 9.1                                      | 1.24| 1.9x10³                         |

1. Lifetimes measured by the B-, G-, and R-OLEDs under current densities of 50 mA/cm² for B- and R-OLEDs and 30 mA/cm² for G-OLED.
2. Current densities of B-, G-, and R-OLEDs for white luminance of 600 cd/m² were determined under 210ppi RGB TE-OLED panel.
3. Accelerating factor

### Table 3. Specifications of the developed 12.3 inch flexible OLED display.

| Size           | 12.3 inch |
|----------------|-----------|
| Resolution     | 1920 x 720 x RGB (167 ppi) |
| Temperature Range | between -40 °C to 95 °C |
| Luminance      | 600 cd/m² |
| Substrate      | Polyimide |

4. Conclusion

The OLED display with long lifetime at 85 °C under the driving of 600 cd/m² white luminance was developed by the optimization of each layer organic material. In particular, new EBL, HBL, and HTL materials were selected for the improvement of the lifetime. We clarified that the charge-carrier balance between the hole and electron inside the EML affects the lifetime of OLEDs. The lifetime becomes long with relatively increasing the ratio of the hole compared with the electron in the EML. The lifetime, which is defined the time for the luminance to decay to 80% of its initial value, shows over 1,000 hours for the B-, G-, and R-OLEDs at 85 °C. With using the new organic materials, we finally fabricated the prototype 12.3 inch flexible OLED display. We expect that the developed OLED display is useful for the automotive display since the lifetime at high temperature is long enough to use this application.

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