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# Highly Efficient and Stable Blue Fluorescent OLED Using Dual EML System

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## Abstract
***In 2022, we presented a high-efficiency improvement for blue fluorescent OLEDs through Dual emitting layer (BH1/BH2 EML) technology to enhance performance. In this study, the light extraction efficiency was improved by controlling the molecular orientation and the spectral shape of blue dopant (BD) in the bule host (BH) on the anode side. Additionally, through mechanism analysis we confirmed that the clear functional separation of the two layers enhanced the Triplet-Triplet Fusion (TTF) efficiency and each BHs deuteration impact resulting in multiplicative extension of lifetime. Furthermore, by applying a dual hole transporting layer (HTL) that contributes to light extraction improvement, bottom-emission (BE) device achieved external quantum efficiency (EQE) =16.3%, and top-emission (TE) device reached BI = 350, LT95@50mA/cm² > 200h.***

## Author Keywords
OLED; Fluorescence; Blue; Triplet-triplet fusion; TTF;

## 1. Introduction
In recent years, Organic Light Emitting Diodes (OLEDs) have rapidly grown in the display market, being widely adopted in applications ranging from small and medium-sized displays like smartphones and PCs to high-end large TVs. Along with this growth, the performance requirements for displays have been increasing year by year. Among the RGB colors, the efficiency of the blue device, which is the only one using fluorescent system, has not be enough. To address this issue, blue phosphorescent and thermally activated delayed fluorescence (TADF) devices have been developed. However, both have not yet reached a practical level in terms of color purity and lifetime [1], [2]. To date, we have had numerous discussions on improving the efficiency of blue fluorescent devices [3], [4], [5]. Most recently, we have presented a Dual emitting layer (EML) technology aimed at achieving the theoretical limit for blue fluorescent devices, and its application in products has already begun [6], [7], [8]. However, we've recognized that there are still existing challenges with blue fluorescent devices utilizing the Dual EML technology, and we believe there is still room for improvement in both efficiency and lifetime. We continue to work on these technical challenges through our ongoing development efforts. This paper reports on the details of these efforts and the results achieved.

## 2. Results and discussions
### 2-1. The challenges of Dual EML technology
Conventional Single EML devices focused on efficiently generating Triplet-Triplet Fusion (TTF) by locally creating excitons within the EML to increase exciton density. However, previous studies have shown that excessive carriers can quench triplet excitons, thereby inhibiting TTF [9], [10]. Additionally, locally increasing the exciton density has a negative impact on device lifetime, making it difficult to balance both efficiency and lifetime. On the other hand, Dual EML devices enable exciton generation across the entire emission layer by utilizing two blue host (BH1/BH2) layers with different triplet energies. By achieving functional separation—recombination occurring in the high triplet energy host (BH1 on the anode side) and TTF in the low triplet energy host (BH2 on the cathode side)—it is possible to generate TTF in a region separate from the recombination zone where excessive carriers exist. This allows for an efficient harvesting delayed emission by TTF, approaching the theoretical limit. Moreover, by dispersing excitons within the EML, this technology also facilitates longer device lifetime simultaneously (Figure 1). Since our previous report, we have made progress in our development, and during this process, we identified two major challenges related to the efficiency of the Dual EML technology. First, we will report on the efforts we have made to address these issues. Then, we will present the results of a more detailed mechanism analysis.

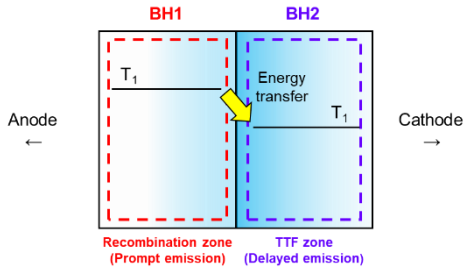


**Figure 1.** Concept of Dual EML system.

### 2-2. Efficiency improvement
We fabricated bottom-emission (BE) and top-emission (TE) devices using a typical device structure, consisting of a Single EML (Single-A) and two types of Dual EML (Dual-A and Dual-B) with different BH1 materials. BH1-A used in Dual-A is the previously developed host material, while BH1-B used in Dual-B is a newly developed material with a different main structure.

**[BE Device] Single-A (x=0), Dual-A and B (x=5):**

ITO(130)/HT-1:HI(10:3%)/HT-1(77.5)/EB-1(7.5)/BH1-X:BD-A(x:2%)/BH2-A:BD-A(20-x:2%)/HB-1(5)/ET-1:Liq(25:33%)/

Liq(1)/Al(80) (nm)

**[TE Device] Single-A (x=), Dual-A and B (x=5):**

Ag/ITO(10)/ HT-1:HI(10:3%)/HT-1(114)/EB-1(7.5)/BH1-X:BD-A(x:2%)/BH2-A:BD-A(20-x:2%)/HB-1(5)/ET-1:Liq(25:33%)/

Yb(1)/Mg:Ag(15:90%)/Cap(65) (nm)

As expected, the devices with Dual EML (Dual-A, B) showed an improvement in the TTF component (delayed emission by TTF), resulting in approximately a 110% increase in EQE compared to the device with Single EML (Single-A) (Table 1, Figure 2). However, it was found that the device using BH1-A (Dual-A) has two major issues. One is a decrease in the prompt component, and the other is an insufficient efficiency improvement in the TE device.


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**Table 1.** Device performance of Single-A, Dual-A and Dual-B at 10 mA/cm$^2$.

| | BE | | | TE |
|---|---|---|---|---|
| | EQE (%) | Prompt (%) | TTF (%) | BI (cd/A/CIE-y) |
| Single-A (Ref.) | 11.2 (100%) | 7.5 | 3.7 | 245 (100%) |
| Dual-A | 12.3 (110%) | 7.2 | 5.1 | 263 (107%) |
| Dual-B | 12.4 (111%) | 7.5 | 4.9 | 271 (110%) |

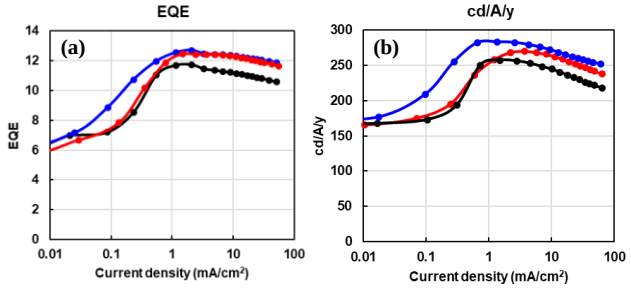


**Figure 2.** (a) Current density vs. EQE and (b) vs. BI (Blue Indices) of Single (Black), Dual-A (Red) and Dual-B (Blue).

### 2-2-1. Control of BD molecular orientation in the BH1

Although the TTF component of Dual-A improved compared to Single-A, the prompt component of Dual-A decreased by 3% compared to Single-A. In order to fully realize the efficiency benefits of the Dual EML technology, it is necessary to restore the prompt component to at least the same level as that of the single emission layer. Mechanistically, the prompt component is primarily considered to originate from emission in the BH1. Moreover, this phenomena are often observed in devices using the previously developed host material for BH1. suggesting that the decrease in the prompt component is likely caused by the BH1. To identify the cause, we fabricated three types of doped films (BH2-A:BD, BH1-A:BD and BH2-B:BD) and conducted an analysis of their emission characteristics.

There was no difference in the photoluminescence quantum yield (PLQY) among the three types of films, but differences in the molecular orientation of BD were observed (Table 2). A brief explanation of the BD orientation evaluation method is as follows. For the angular dependence of the emission, the measured data were obtained using the orientation measurement device IMS-5000 (Asahi Bunkou), and the calculated data were acquired using the optical simulator Setfos (Fluxim). The closest Pz component between the measured and calculated values was determined using a Python program, and 1 - Pz = S' was calculated.

As a result, the orientation of BD-A in BH1-A was found to be 3% lower than that of BD-A in BH2-A, and this decrease corresponds to the decline in the prompt component. Compared to the Single-EML, the decrease in the prompt component in the Dual EML was found to be due to the reduced BD orientation in the BH1 layer (Table 2). On the other hand, in BH1-B, which has a newly developed material, no decrease in the prompt component or reduction in BD orientation was observed. By changing the main structure of BH1, we successfully controlled the orientation of BD. It is believed that the orientation of BD in the BH1 is largely influenced by the main structure of BH1. However, through the acquisition of numerous experimental data, we have found that it is not solely determined by the main structure of BH1. Instead, the combination of both the BH1 and BD plays a significant role in determining the orientation.

**Table 2.** Prompt Efficiency (BE) of Single and Dual EML at 10 mA/cm$^2$ and Characteristics of doped Film.

| | BE | Film | |
|---|---|---|---|
| | Prompt (%) | PLQY | S' |
| Single-A (Ref.) | 7.5 (100%) | 100% | 0.950 (100%) |
| Dual-A | 7.2 (97%) | 100% | 0.925 (97%) |
| Dual-B | 7.5 (100%) | 100% | 0.950 (100%) |

### 2-2-2. Control of BD spectra in the BH1

The second challenge is that the efficiency improvement of the Dual-A TE device was 107%, which is smaller than the 110% improvement seen in the BE device (Table 3). Since the EQE of the TE device shows a similar improvement to that of the BE device, the efficiency drop from the BE to the TE device is believed to be due to a loss in light extraction efficiency. To investigate this, we measured the photoluminescence (PL) spectra of three doped films (BH2-A:BD, BH1-A:BD and BH2-B:BD) (Figure 3).

**Table 3.** BI (TE) of Single and Dual EML at 10 mA/cm$^2$ and Characteristics of doped Film.

| | TE | | Film | |
|---|---|---|---|---|
| | BI (cd/A/CIE-y) | EQE (%) | λmax (nm) | FWHM (nm) |
| Single-A (Ref.) | 245 (100%) | 22.1 (100%) | 462 | 20 |
| Dual-A | 263 (107%) | 24.3 (110%) | 463 | 21 |
| Dual-B | 271 (110%) | 24.8 (111%) | 463 | 20 |

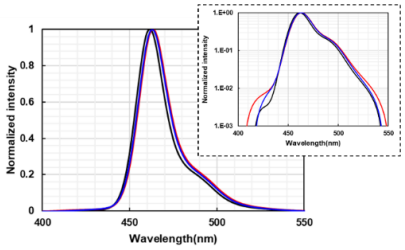


**Figure 3.** Normalized PL spectra of Single (Black), Dual-A (Red) and Dual-B (Blue) at 10mA/cm$^2$.

As a result, compared to BD-A emission in BH2-A, the PL spectrum of BD-A in BH1-A showed a redshift in the peak wavelength and a broadening of the full width at half maximum (FWHM). This indicates that the spectral changes of BD-A in the BH1-A are the cause of the loss in light extraction efficiency in the TE device. On the other hand, in the PL spectrum of BD-A in BH1-B, the FWHM remained unchanged, and only the redshift in the peak wavelength was observed. This allowed us to limit the efficiency drop in the TE device to around 1%. By changing the main structure of BH1, we successfully controlled the spectrum of

BD and suppressed the light extraction efficiency loss in the TE device. However, even with BH1-B, full control has not yet been achieved, and further adjustments beyond the main structure are necessary. These aspects will also be reported on the day of the presentation.

### 2-3. Mechanism analysis

Next, we will report the results of the newly mechanism analysis for Dual EML technology. By applying this technology, it is possible to harvest TTF emission effectively. However, it has been observed that the effect varies depending on the BH1 characteristics. In the previous report, we utilized the spectral change of BD in BH1 and calculated the emission ratio in a simplified method. We found that when the emission ratio was closer to the ideal state (62.5% of the emission from singlet excitons (Prompt) in BH1 and 37.5% from triplet excitons (TTF) in BH2), we got the highest TTF efficiency. However, this method could not observe origin of emission from singlet excitons (Prompt) in BH1 and triplet excitons (TTF) in BH2, as per the concept. In this study, we evaluated the BE device using different color dopants: BD on the BH1 and green dopant (GD) on the BH2 (Figure 4). By using an optical filter (525 nm, long pass), we could isolate the GD emission from the BH2, allowing us to track whether the emission originated from the Prompt or TTF component. Furthermore, we hypothesized that by comparing the three types of devices—Single EML, Dual EML with low TTF, and Dual EML with high TTF—we could observe differences in the emission components.

As in the previous case, we fabricated BE devices with a typical device structure: Single EML(Single-A) and two types of Dual EML (Dual-C, D) with different BH1 materials. Additionally, we fabricated devices with GD-A doped on the cathode side of Single EML or the BH2 side of Dual EML. The BH1-C and BH1-D materials used in the Dual EML devices also have newly developed materials with a different main structure. Furthermore, GD-A was chosen to suppress energy transfer from BD-A by selecting a material with a small Stokes shift (i.e., minimal overlap between the fluorescence spectrum of BD-A and the absorption spectrum of GD-A) and a T1(=2.0eV) suitable for the Dual EML concept.

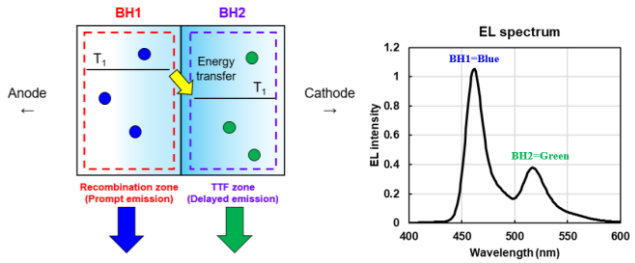


**Figure 4.** Mechanism analysis device of Dual EML (B/G) and EL spectrum.

**[BE Device (B/B)] Single-A (x=0), Dual-C and D (x=10):**

ITO(130)/HT-1:HI(10:3%)/HT-1(77.5)/EB-1(7.5)/BH1:BD-A(x:2%)/BH2-A:BD-A(20-x:2%)/HB-1(5)/ET1-1:Liq(25:33%)/Liq(1)/Al(80) (nm)

**[BE Device (B/G)] Single-A:**

ITO(130)/HT-1:HI(10:3%)/HT-1(77.5)/EB-1(7.5)/BH2-A:BD-A(10:2%)/BH2-A:GD-A(10:2%)/HB-1(5)/ET1-1:Liq(25:33%)/Liq(1)/Al(80) (nm)

**[BE Device (B/G)] Dual-C and D:**

ITO(130)/HT-1:HI(10:3%)/HT-1(77.5)/EB-1(7.5)/BH1-X:BD-A(10:2%)/BH2-A:GD-A(10:2%)/HB-1(5)/ET1-1:Liq(25:33%)/Liq(1)/Al(80) (nm)

The results for the blue device (B/B) showed that the Prompt component was similar across all three devices, with differences only observed in the TTF component. To further analyze this, we fabricated blue-green devices (B/G) and performed transient EL measurements using an optical filter (525 nm, long pass) to isolate GD emission only (Table 4).

In the Single EML device, the GD emission on the cathode side was weak and TTF emission couldn't be detected. This result suggests that, in the Single EML device, only a small amount of emission is contributed from the cathode side, with the majority of the emission coming from the anode side. In contrast, GD emission from the BH2 side of both Dual EMLs was detected, and the TTF ratio increased. This confirms that, in the Dual EML device, the BH2 side contributes to the emission from triplet excitons (TTF) as per the concept. In particular, for the Dual-D showed high TTF, the TTF ratio reached 100%, demonstrating that the BH2 side contributes only to the TTF component. In the case of the Dual-C showed lower TTF, the TTF ratio was 60%, indicating that the BH2 side also contributed to the Prompt component.

This analysis proves that Dual EML system is functioning according to the concept and highlights that maximizing TTF efficiency in the Dual EML requires achieving an ideally fully functional separation of the BH1 and BH2 components.

**Table 4.** Device performance (B/B and B/G) of Single-A, Dual-C and Dual-D at 10 mA/cm$^2$.

| | BE (B/B) | | | | BE (B/G)* |
|---|---|---|---|---|---|
| | EQE (%) | Prompt (%) | TTF (%) | TTF ratio | TTF ratio |
| Single-A (Ref.) | 11.2 (100%) | 7.5 | 3.7 | 0.34 | Not Detect |
| Dual-C | 11.9 (107%) | 7.5 | 4.4 | 0.37 | 0.60 |
| Dual-D | 12.4 (111%) | 7.5 | 4.9 | 0.39 | 1.00 |

* using an optical filter (525 nm, long pass)

### 2-4. Lifetime improvement

Deuterated materials have been utilized as a method to enhance the lifetime of OLED devices. The target of deuteration is primarily the EML, but the combined effects of this approach have not been thoroughly investigated. In particular, since the Dual EML system adds an extra layer and the emission mechanism changes, we investigated the impact of deuteration of BH materials on the lifetime improvement in both Single and Dual EML.

Using the deuterated materials of BH2-A from Single EML device (Single-A) and BH1-D and BH2-A from Dual EML device (Dual-D), we fabricated BE devices. As a result, the deuterated Single EML exhibited long lifetime of approximately 160%. On the other hand, the deuterated Dual EML showed long lifetime of 228% due to the cumulative effect of deuterated BH1 and BH2, demonstrating a greater lifetime improvement compared to the Single EML (Table 5). Regarding the difference in effect, we believe the difference in

emission mechanisms is involved. As described in section 2-3, in the Single EML device, only the anode side contributes to the emission. In contrast, in the Dual EML device, due to the functional separation, both the anode side of BH1 and the cathode side of BH2 contribute to the emission. Since the emission-contributing area is expanding in the Dual EML, the lifetime improvement effect from deuteration was more pronounced. Currently, using section 2-3 devices, we are conducting lifetime analysis and will report the results on the day.

**Table 5.** Efficiency of deuterated Single EML and Dual EML devices for long lifetime at 50mA/2/cm$^2$.

| | D Layer | BH1 | BH2 | LT95 (h) | Relative LT95 |
|---|---|---|---|---|---|
| Single-A | 0 | | H | 60 | 100% |
| | 1 | | D | 96 | 160% |
| Dual-D | 0 | H | H | 87 | 100% (145%) |
| | 1 | D | H | 125 | 144% (209%) |
| | 1 | H | D | 157 | 180% (261%) |
| | 2 | D | D | 198 | 228% (331%) |

() is Relative to Single-A (D layer=0)

### 2-5. Current achieved performance

Building on the content reported so far, we fabricated a device (Dual-E) with Dual EML, applying Dual Hole Transporting Layer (HTL) with high-N and low-N materials to improve light outcoupling efficiency. As a result, we achieved a very high-efficiency BE device with an EQE = 16.3%. Using a similar structure, we also fabricated a TE device, which demonstrated a BI = 350 and LT95@50mA/cm² of over 200 hours (Table 6, Figure 5).

As the importance of new high-efficiency technologies for blue devices continues to rise, we believe that it is possible to achieve comparable efficiency with blue fluorescent devices while simultaneously ensuring long lifetimes. Today we've reported BI = 350 efficiency, however we believe our material and device design strategy will allow to pull out the full potential of fluorescent Blue emission.

**[BE Device] Dual-E (*Deuterated material):**

ITO(130)/HT-2:HI(10:3%)/HT-2(37.5)/HT-3(40)/EB-2(7.5)/

BH1-D*:BD-A(5:2%)/BH2-A*:BH2-B*:BD-A(15:40%;2%)/

HB-2(5)/ET1-1:Liq(25:33%)/Liq(1)/Al(80) (nm)

**[TE Device] Dual-E (*Deuterated material):**

Ag/ITO(10)/ HT-2:HI(10:3%)/HT-2(61)/HT-3(50)/EB-2(7.5)/
BH1-D*:BD-A(5:2%)/BH2-A*:BH2-B*:BD-A(15:40%;2%)/

HB-2(5)/ET-1:Liq(25:33%)/Yb(1)/Mg:Ag(15:90%)/Cap(65) (nm)

**Table 6.** Device performance of Dual-E at 10 mA/cm$^2$. Efficiency of BE is*LT95 was measured at 50mA/cm$^2$.

| | | Voltage (V) | CIE x | CIE y | EQE (%) | BI (cd/A/CIE-y) | LT95* (h) |
|---|---|---|---|---|---|---|---|
| Dual-E | BE | 3.46 | 0.134 | 0.067 | 16.3 | 156 | >200 |
| | TE | **3.58** | **0.139** | **0.042** | 32.8 | 350 | **>200** |

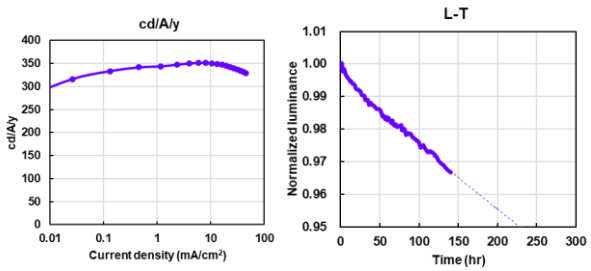


**Figure 5.** Current density vs. BI and Luminance decay of Dual-E (TE device).

## 3. Conclusion

We have developed Dual EML technology as a high-efficiency approach for blue fluorescent devices. This time, we have successfully advanced Dual EML technology to an even higher level in terms of both efficiency and lifetime. In terms of efficiency, we succeeded in suppressing the efficiency decrease in the BH1 on the anode side by controlling the orientation factor and spectrum shape of the BD. Additionally, new mechanism analyses revealed that clear functional separation in Dual EML system not only maximizes TTF efficiency but also enhances the long-lifetime effect through the deuteration of BH. Furthermore, by combining the Dual EML technology with Dual HT for improved light outcoupling efficiency, we achieved a highly successful balance of both efficiency and lifetime: an EQE = 16.3% for the BE device, and a BI = 350 with LT95@50mA/cm² > 200h for the TE device.

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