Letter

Development of White Light Emitting Diodes by Multi-layered Red, Green, and Blue Phosphors Excited by Near-ultraviolet Light Emitting Diodes


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ABSTRACT

Cascade excitation process and cascade excitation loss (absorption loss) have been investigated by bilayer (BL-) LEDs. The cascade excitation loss was suppressed by multi-layered (ML-) LED structure, in which red phosphor is placed at the bottom. We have also developed white LEDs by using a ML-structure of red, green, and blue phosphors for the purpose of reducing the cascade excitation. Luminous flux of ML-white LEDs showed higher value under low correlation color temperature (CCT=2813K) and high color-rendering index (Ra=94) conditions than conventional phosphor mixed-white LEDs. The angular distribution of chromaticity in the ML-R/G/B white LED was comparable to that in mixed-RGB white LED and was very small.

KEYWORDS: near-ultraviolet LED, phosphor conversion white LED, cascade excitation, luminous efficacy

1. Introduction

LED is a promising candidate as a novel light source of 21st century for their high efficiency, long lifetime, Mercury-free, etc. resulting energy saving and environmental safeguards. LEDs are used for a backlight of a small display such as cellular phone and a partial illumination. To extend LED application to general lighting, higher efficiency and luminous flux of white LED is required. Addition to the high efficiency and high flux, white LEDs for general lighting need to have high color rendering (suitable value for comfortable living) and uniform light distribution under wide space (Lambertian and no dispersion of color). It is difficult to satisfy those requirements at the same time.

White LEDs using complementary white such as blue-LED and YAG yellow phosphor system generally show high efficiency but inhomogeneous distribution and poor color rendering. It is unsuitable for general lighting, and many efforts have been performed to solve those problems. On the other hand, near-ultraviolet (n-UV) LED and multi-phosphor system, such as red, green and blue (RGB) phosphor system, has advantages on uniformity of light distribution resulting from the characterization of phosphor-converted light and on high color rendering. The efficiency of n-UV LED and RGB phosphor system is, however, inferior to that of complementary system, because of relatively low efficiency of n-UV LED chip and cascade excitation effect as shown in Fig.1, which appears in multi-phosphor system to get high color-rendering. Cascade excitation causes "cascade excitation loss" due to external quantum efficiency of phosphor.

Generally, the phosphors used for n-UV excited phosphor-conversion white LEDs are a mixture of red, green and blue phosphors. In contrast, we have investigated multi-layered phosphor structures for the purpose of control of cascade excitation loss, and have demonstrated multi-layered phosphor-conversion white LED with high efficiency and high color-rendering.

![Figure 1: Schematic image of cascade excitation in the case of red, green and blue phosphor excited by n-UV light](image)

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2. Experimental procedures

Surface-mount type LEDs, which were directly bonded on ceramic packages without sub-mount (we call them direct flip-chip bonding technique: DFCB), were used as the n-UV excitation source. Dimethyl silicone resin was used as the binder of phosphors. Multi-layered (ML) phosphor structures were fabricated as follows; Dimethyl silicone resin, in which red, green and blue phosphors for ML-phosphors were individually bound, was dispensed on NF/R/UV LED packages. They were then incompletely cured for each phosphor layer to avoid the formation of mixed boundary and to keep adhesive strength between each phosphor layer. LEDs encapsulated by mixed phosphor were also fabricated in comparison.

At first, cascade excitation loss was estimated by using simple structures consisting of red and blue phosphors as shown in Fig. 2. The structure was bilayer, of which the top layer was blue phosphor and the bottom layer was red phosphor (BL-R/B). The second structure was the inverted bilayer (BL-B/R). The third structure was monolayer of mixed red and blue phosphor (Mixed RB). The amount of R and B phosphors and that of dimethyl silicone resin in the BL-R/B, the BL-B/R and the mixed RB phosphor-conversion LEDs were controlled to be same. If cascade excitation loss exists in this system, BL-R/B suppresses the energy loss by absorption but BL-B/R enhances the energy loss.

Then, white LEDs by multilayers, of which the top layer was blue phosphor, the middle layer was green phosphor, and the bottom layer was red phosphor, was fabricated (ML-RG/B). White LED by a monolayer of Mixed red, green, and blue phosphor was also fabricated (Mixed-RGB). The amount of red, green and blue phosphors and that of dimethyl silicone resin in both white LEDs were also controlled to be same.

Radiant flux, luminous flux, correlation color temperature (CCT) and general color-rendering index (Ra) were measured by integrating sphere.

![Figure 2: Packaging model of BL/R/B, mixed RB, BL/R/B](image)

3. Results and discussion

Radiant fluxes of red and blue components in the spectra were individually evaluated for the BL-R/B, the BL-B/R and the mixed-RB LED as shown in Table 1. The radiant flux of red component was higher in the BL-B/R and the mixed-RB LED. On the other hand, the radiant flux of blue component was higher in the BL-R/B LED. These results indicated that red phosphor in the BL-B/R or the mixed-RB LED absorbed blue emission from the underlying blue phosphor or adjacent blue phosphor and that red phosphor in the BL-R/B LED didn’t absorb blue emission from the overhead blue phosphor. These tendencies are also understood by comparing the radiant flux of blue/the radiant flux of red (BR ratio), which was the highest in the BL-R/B LED. In addition, the sum of the radiant flux of red and blue components was highest in BL-R/B LED. This is considered that energy loss exists in re-absorption processes and that the energy loss increases with increase of re-absorption probability in the LED package, which is related to the distance and/or placement of blue and red phosphor. It is consistent with the behavior of radiant flux of red and blue components. To suppress the cascade excitation, phosphor with the lowest energy of excitation energy must be placed at the bottom layer of ML-LED.

<table>
<thead>
<tr>
<th>Table 1: Comparison of radiant flux in RB system</th>
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<tbody>
<tr>
<td>Radiant flux [mW]</td>
</tr>
<tr>
<td>R</td>
</tr>
<tr>
<td>---</td>
</tr>
<tr>
<td>BL-R/B</td>
</tr>
<tr>
<td>Mixed-RB</td>
</tr>
<tr>
<td>BL-B/R</td>
</tr>
</tbody>
</table>

Next, the characteristics of ML-RG/B and mixed-RGB white LEDs were compared. The radiant flux of red, green, blue and their sum are shown in Table 2. The radiant flux of red component in the ML-RG/B LED was much larger than that in the mixed-RGB LED. In contrast, radiant fluxes of blue and green components in the ML-RG/B LED were smaller than those in the mixed-RGB LED. The difference of these tendencies between the ML-RG/B and the mixed-RGB is understood as the result of the placement difference of phosphors; i.e. in the ML-RG/B LED, n-UV light from LED was first absorbed by the red phosphor at the bottom layer, but exponentially drops at the middle green layer and the top blue layer; on the other hand, all phosphor were equivalently excited by the n-UV LED in the mixed-RGB LED.

The sum of radiant fluxes was higher in the ML-RG/B LED. This is also due to the existence of cascade excitation loss as well as the case of BL-R/B and mixed-RB LED. These results imply that the optimized ML white LED has large advantage in efficiency especially at low CCT region.

Table 3 shows the luminous flux, CCT, and Ra of two white LEDs. In spite of low color temperature (about 2600 K), high Ra and high luminous flux is simultaneously obtained. This also supports above expectation.

At last, the angular dependence of chromaticity (x, y) of the ML-RG/B white LED, the mixed-RGB white LED and cannonball-shaped Blue-YAG white LED were
Table 2 Comparison of radiant flux in RGB

<table>
<thead>
<tr>
<th></th>
<th>R [mW]</th>
<th>G [mW]</th>
<th>B [mW]</th>
<th>sum [mW]</th>
</tr>
</thead>
<tbody>
<tr>
<td>ML-R/ G/B</td>
<td>19.97</td>
<td>7.72</td>
<td>0.93</td>
<td>28.63</td>
</tr>
<tr>
<td>Mixed-RGB</td>
<td>10.57</td>
<td>10.81</td>
<td>1.46</td>
<td>22.85</td>
</tr>
</tbody>
</table>

Table 3 Characteristics (lumens, CCT, Ra) of the ML-R/ G/B and the mixed-RGB white LEDs

<table>
<thead>
<tr>
<th>Luminous flux [lm]</th>
<th>CCT [K]</th>
<th>Ra</th>
</tr>
</thead>
<tbody>
<tr>
<td>ML-R/ G/B</td>
<td>8.22</td>
<td>2613</td>
</tr>
<tr>
<td>Mixed-RGB</td>
<td>7.82</td>
<td>4375</td>
</tr>
</tbody>
</table>

Figure 3 Angular dependences of chromaticity for blue-YAG LED, mixed-white LED and ML-white LED

4. Summary

We have investigated the ML-R/ G/B LED structure by using DFCB n-UV LED as the excitation source. Cascade excitation process and cascade excitation loss (absorption loss) existed in multi phosphor system; however, the loss was suppressed by ML-LED structure. In ML-LED, the bottom phosphor layer is preferentially excited, and it is desirable to suppress cascade excitation loss that red phosphor is placed at the bottom. Consequently, it is suited for ML-LED to obtain high-efficiency in low CCT.

The angular distribution of chromaticity in the ML-R/ G/B white LED was comparable to that in mixed-RGB white LED and was very small. The ML-R/ G/B white LED is one of the solutions of general light source.

Acknowledgement

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References


