

White ACPEL Device with ZnS:Cu,Cl, Tb₃Al₅O₁₂:Ce³⁺, and CaS:Eu²⁺ Phosphors Using a Layered Structure

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ABSTRACT—Improvement of the color rendering index (CRI) and luminance of a white alternate current powder electroluminescent (ACPEL) device has been attempted using ZnS:Cu,Cl, Tb₃Al₅O₁₂:Ce (TAG:Ce), and CaS:Eu phosphors with a layered structure. The device with TAG:Ce and ZnS:Cu,Cl phosphors showed a CRI of 75, with a luminance increase of about 30% depending on the thickness of the TAG:Ce. Further CRI improvement was attempted using CaS:Eu. When they were separately screen-printed, the CRI was increased up to 89 with no decrease in luminance.

Keywords—ZnS:Cu,Cl, TAG:Ce, CaS:Eu, ACPEL, CRI.

I. Introduction

Alternate current powder electroluminescent (ACPEL) devices have been studied since the 1970s due to their several advantages: low power consumption, uniform light emission within a large area, and their easy screen-print fabrication method. Moreover, if plastic substrates are used, they have further advantages such as low weight, compactness, and robustness. However, due to their low brightness and short lifetime, their application is relatively limited to signage and backlights for liquid crystal displays (LCDs), cellular phones, and personal digital assistants (PDAs) [1], [2].

For such applications, white light is the most important

aspect because ACPEL devices are usually used as backlight units. However, conventional white ACPEL devices have suffered from relatively low luminance and poor CRI characteristics due to their limited phosphor performance. Current fabrication methods and their characteristics for white ACPEL devices are the following:

- the blending of a blue-green emitting ACPEL ZnS:Cu,Cl phosphor with Y₃Al₅O₁₂:Ce (YAG:Ce), where the CRI is less than 70
- the blending of a blue-green emitting ACPEL ZnS:Cu,Cl phosphor with Ca₂Si₅N₈:Eu phosphor, where the CRI is 75 to 85 while the luminance decreases by about 30% to 40% as compared with that of blue devices from ZnS:Cu,Cl
- a single ZnS:Mn,Cu,Cl ACPEL phosphor with bluish-green and orange emission, where the CRI is not very good due to the orange emission by Mn, and the luminance is relatively low [3]–[5]

Recently, TAG:Ce phosphor has been reported to overcome the CRI issue of YAG:Ce in white LED applications [6]. According to the report, TAG:Ce phosphor has a more intense emission than YAG:Ce phosphor in the red spectral region, and the CRI characteristics of white light is improved. Therefore, in this letter, we propose a way to improve the luminance and CRI of a white ACPEL device by a structural change in an ACPEL device and with TAG:Ce and CaS:Eu phosphor, respectively.

II. Experiment

The main EL phosphor was a commercial ZnS:Cu,Cl synthesized by ELK (Korea). TAG:Ce was synthesized using a solid-state reaction method [6]. However, CaS:Eu was synthesized using a modified solid-state reaction that uses a quartz ampule to avoid oxidation of CaS [7]. An ACPEL

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device was fabricated using a conventional screen-printing method [8]. The dielectric and rear electrode materials are BaTiO₃ paste and graphite paste, respectively. Photoluminescence (PL) and PL excitation (PLE) were characterized using a DARSA PRO 5100 PL system (Professional Scientific Instrument Co., Korea) at room temperature. The EL spectrum was recorded using optic fiber and the DARSA PRO system. Luminance was measured using a BM-7 Luminance colorimeter (TOPCON, Japan).

III. Results and Discussion

1. White ACPEL Device Using ZnS:Cu,Cl and TAG:Ce Phosphors

Figure 1(a) shows the PL and PLE spectra of TAG:Ce and an EL spectrum of ZnS:Cu,Cl. As shown in Fig. 1, excitation and emission peaks are located at almost the same wavelength of about 455 nm, which means that the excitation of TAG:Ce by the emission of ZnS:Cu,Cl must be very effective. Figure 1(b) shows an SEM image of a ZnS:Cu,Cl ACPEL device.

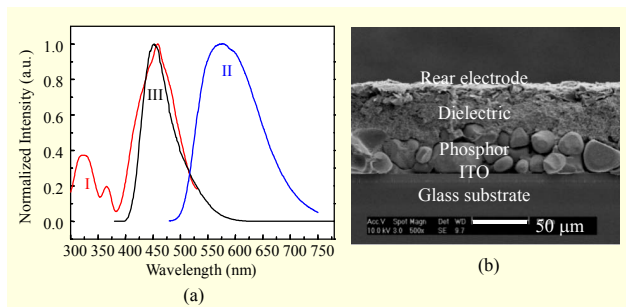


Fig. 1. (a) I: PLE spectrum of TAG:Ce ($\lambda_{em}=560$ nm); II: PL spectrum of TAG:Ce ($\lambda_{ex}=450$ nm); III: EL spectrum of ZnS:Cu,Cl with 100 V at 400 Hz, and (b) SEM image of conventional ACPEL device.

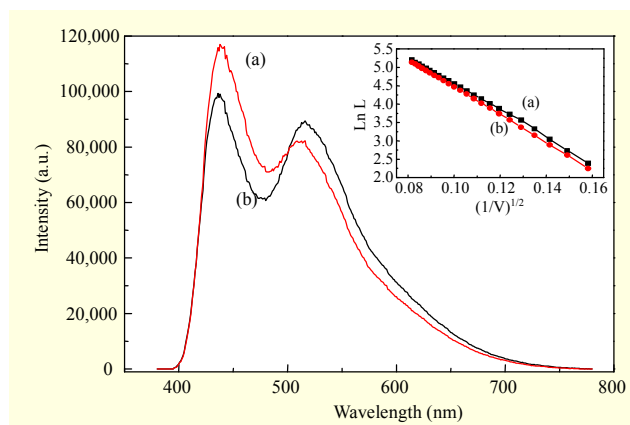


Fig. 2. EL spectra of white devices with (a) TAG:Ce (5 layers) and (b) TAG:Ce (7 layers). The right inset shows the relationship between the luminance (L) and applied voltage.

The TAG:Ce layer was screen-printed on top of the glass substrate of blue ACPEL devices. Devices screen-printed five and seven times with TAG:Ce showed 96 cd/m² and 87 cd/m² with 100 V at 400 Hz, respectively, and luminous efficacy of 3.94 lm/W and 3.61 lm/W, respectively. Their CIE color coordinates were (0.248, 0.429) and (0.227, 0.391), both of which were bluish-white in color, and their CRIs were 72 and 75, respectively.

Figure 2 shows the EL spectra of two white ACPEL devices. The yellow emission increases and the blue emission decreases as the thickness of the TAG:Ce layer is increased. That is to say, more blue light is absorbed by the TAG:Ce as the amount increases. However, the blue and yellow emission peaks of white ACPEL devices are located at about 430 nm and 510 nm, respectively. Those emissions are blue-shifted compared to the characteristic 450 nm blue emission of ZnS:Cu,Cl and the 550 nm yellow emission of TAG:Ce. The blue-shift of ZnS:Cu,Cl is probably due to the decrease of 450 nm blue light by the absorption of TAG:Ce, and the blue-shift of TAG:Ce is due to the addition of ZnS:Cu,Cl emission to the TAG:Ce emission in the tail region of the ZnS:Cu,Cl emission.

2. CRI Improvement Using ZnS:Cu,Cl, TAG:Ce, and CaS:Eu Phosphors

When ZnS:Cu,Cl and TAG:Ce phosphors are used, the CRI does not show as much improvement as expected. Therefore, CaS:Eu, which absorbs blue light and emits red light, was added to improve the CRI of the white ACPEL devices. The devices were fabricated using two different methods.

A. Mixture of CaS:Eu and TAG:Ce PL Phosphor Layer/ITO Glass/ZnS:Cu, Cl EL Phosphor Layer/BaTiO₃ Insulating Layer/Rear Electrode Layer

The mixture of CaS:Eu and TAG:Ce at a ratio of 2:1 (in wt%) was screen-printed seven times on top of ITO glass. The device showed 86.8 cd/m² with 100 V at 400 Hz, which is slightly lower than that of a device without CaS:Eu phosphor. However, the CIE color coordinate was (0.218, 0.342), which is almost that of blue light. This means that CaS:Eu phosphor does not efficiently improve the CRI in this method.

Figure 3 shows EL spectra of a device without CaS:Eu phosphor and device with a mixture of CaS:Eu and TAG:Ce phosphors. As shown in Fig. 3, the yellow emission of the device with the CaS:Eu and TAG:Ce mixture disappears. This is likely the main reason for blue light emission instead of white light emission. The disappearance of the yellow emission is due to the absorption of yellow light by CaS:Eu because the absorption band of CaS:Eu is overlapped with the yellow emission band of TAG:Ce as shown in the inset of Fig. 3.

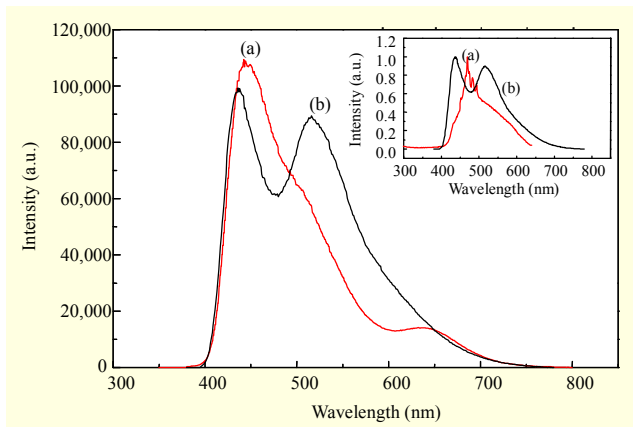


Fig. 3. EL spectra of white devices (a) with and (b) without CaS:Eu. The right inset shows a (a) PLE spectrum of CaS:Eu and (b) EL spectrum of a white device.

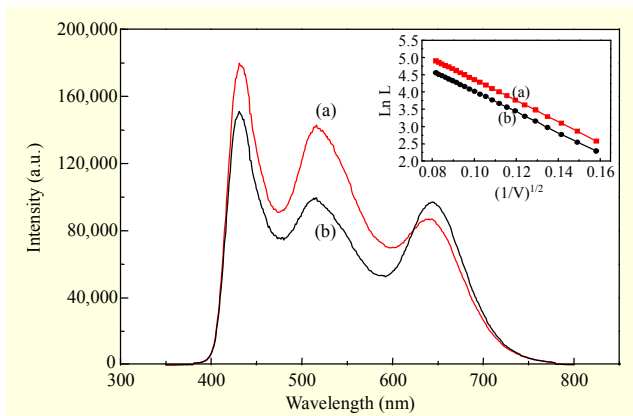


Fig. 4. EL spectra of white devices with (a) CaS:Eu (1 layer) and (b) CaS:Eu (2 layers). The right inset shows the relationship between luminance (L) and applied voltage.

Thus, even though the blue EL emission of ZnS:Cu,Cl excites TAG:Ce, generating yellow light, the yellow light is reabsorbed by CaS:Eu.

B. TAG:Ce PL Phosphor Layer/CaS:Eu PL Phosphor Layer/ITO Glass/ZnS:Cu,Cl EL Phosphor Layer/BaTiO₃ Insulating Layer/Rear Electrode Layer

As previously mentioned, due to the absorption of yellow light by CaS:Eu, white light was not generated. To overcome the re-absorption of yellow light, the CaS:Eu layer was screen-printed separately from the TAG:Ce layer. If TAG:Ce is excited later than CaS:Eu, yellow light should not be absorbed by CaS:Eu. Therefore, CaS:Eu was first screen-printed on top of ITO glass and TAG:Ce was screen-printed later. As a result, devices screen-printed one and two times showed 78.5 cd/m² and 56 cd/m² with 100 V at 400 Hz, respectively, and luminous efficacy of 3.32 lm/W and 2.53 lm/W, respectively. Generally, the brightness of TAG:Ce is much stronger than that of CaS:Eu

when they are excited by blue light. In this method, blue light from ZnS:Cu,Cl excited CaS:Eu first, and the remainder of the blue light excited TAG:Ce later. Thus, TAG:Ce was not efficiently excited, and the total luminance of the white device was decreased compared to that of the white device generated by only TAG:Ce. However, unlike the conventional white ACPEL devices, luminance still remains almost the same as that of the blue device in this method, even though the CRI is improved up to 89.

EL spectra were measured to confirm blue, yellow, and red emissions. As shown in Fig. 4, yellow light is not absorbed by CaS:Eu, and there are 430 nm blue, 516 nm yellow, and 644 nm red emissions. Thus, CaS:Eu is proven to work efficiently in improving the CRI of a white ACPEL device.

IV. Conclusion

A white ACPEL device was fabricated using blue EL emitting ZnS:Cu,Cl and yellow PL emitting TAG:Ce phosphors with a layered structure to improve brightness and CRI. The luminance decreases and CRI increases with an increase of TAG:Ce because more blue light is absorbed by the TAG:Ce yellow phosphor. Further CRI improvement was achieved using a red PL emitting CaS:Eu phosphor with TAG:Ce phosphor. When they are separately screen-printed, CRI is improved up to 89, while the luminance does not decrease compared with that of the blue device.

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