

# Eutectic Phosphor for High-Power Excitation

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## ABSTRACT

*High-power blue-emitting laser diodes have been applied to projectors and lighting systems as excitation sources for yellow-emitting phosphors. These phosphors must be robust and should not exhibit thermal quenching. This work developed durable eutectic phosphors with a thermal conductivity of above 20 W/m·K.*

## 1 Introduction

Solid-state lighting devices are widely used as general light sources and for projectors and backlights in liquid crystal displays. These units typically consist of yellow-emitting Ce:Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub> (Ce:YAG) phosphors and blue light emitting diodes (LEDs) acting as excitation sources for the phosphor and cover blue component. Recent advances in this field have led to the use of laser diodes (LDs) as excitation sources, as a means of increasing the light output of projectors, outdoor lighting systems, headlights and so on. However, the phosphors used for LEDs are often not applicable to LDs. The excitation density for LDs is very high due to their greater power consumption and small spot sizes. As such, phosphors are exposed to high power levels and tend to rapidly degrade. Even in the case that the phosphor does not degrade, its temperature is significantly increased such that thermal quenching occurs.

A rotating phosphor wheel that reduces the excitation density has been proposed as a means of mitigating this problem [1]. Unfortunately, it is challenging to design compact versions of such systems and there are also issues related to noise and reliability because a drive unit is necessary. Therefore, a fixed phosphor light source would be preferable to a rotating wheel. In this case, it would be desirable to employ a phosphor that does not degrade even in response to high-power excitation and also does not undergo quenching at elevated temperatures.

The present study demonstrates a new type of phosphor that exhibits superior durability in response to high-power excitation, comprising a eutectic mixture of Ce:YAG and sapphire [2-3]. The basic characteristics of this material, which we term EPOCH-Neo, were evaluated and experimental devices made of the eutectic phosphor with a heat sink were fabricated. The phosphor was also coated with several layers, including an Al or Ag reflective

layer, and affixed to a heat sink with solder. Devices having various phosphor thicknesses were examined under different conditions.

## 2 Basic characteristics of EPOCH-Neo

Figure 1 presents the luminescence spectrum obtained from EPOCH-Neo having a Ce concentration of 0.5mol% with excitation at 450 nm as acquired using a Jasco FP-8550 instrument. The internal quantum efficiency, external quantum efficiency and absorption ratio for this material were 88%, 77% and 87% respectively.

Figure 2 shows a scanning electron microscopy (SEM) image of EPOCH-Neo. This material was a melt-grown composite in which Ce:YAG and sapphire were homogeneously intermixed without the use of an organic binder and without air spaces or boundary layers.

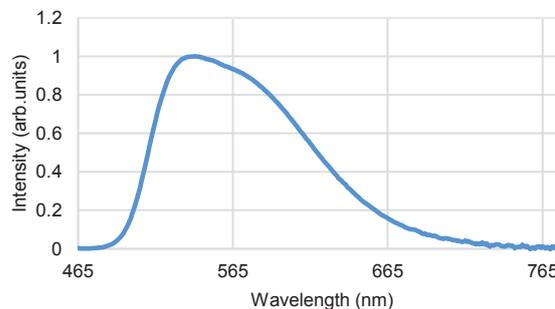


Fig. 1 Luminescence spectrum of EPOCH-Neo specimen.

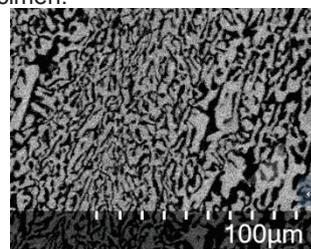


Fig. 2 SEM image of EPOCH-Neo specimen in which bright and dark areas correspond to Ce:YAG and sapphire, respectively.

In EPOCH-Neo, heat generated as a result of a Stokes shift or non-radiative transitions is dissipated along the Al<sub>2</sub>O<sub>3</sub> regions because this oxide has a higher

thermal conductivity ( $>20$  W/m·K) than that of Ce:YAG. In the present study, this material was synthesized using an edge-defined film-fed growth method that tended to generate plate-like chips.

The effects of temperature on the quenching characteristics of both EPOCH-Neo and pure Ce:YAG powder were assessed. In these trials, the sample was heated from 25 to 245 °C and luminescence spectra were acquired at 10 °C intervals. The spectra were subsequently integrated over the wavelength range of 465 to 780 nm and these values were plotted as percentages, with the value at 25 °C set to 100% (Fig. 3). The intensity of the luminescence produced by the Ce:YAG powder was found to be reduced by approximately 30% at 245 °C.

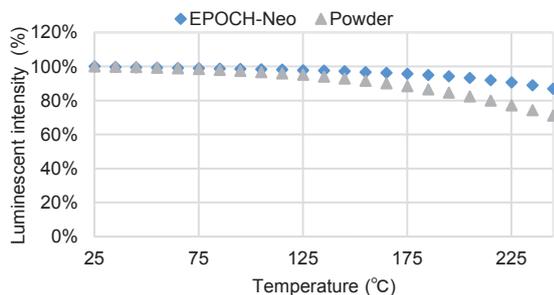


Fig. 3 Thermal quenching characteristics of EPOCH-Neo and Ce:YAG powder.

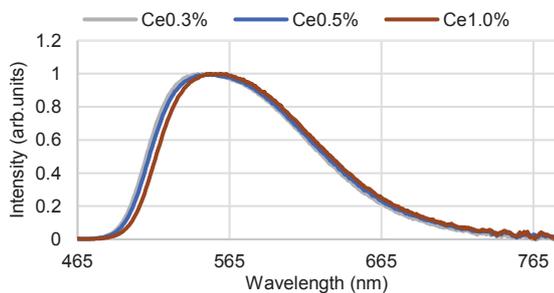


Fig. 4 Luminescence spectra of EPOCH-Neo samples with varying Ce content.

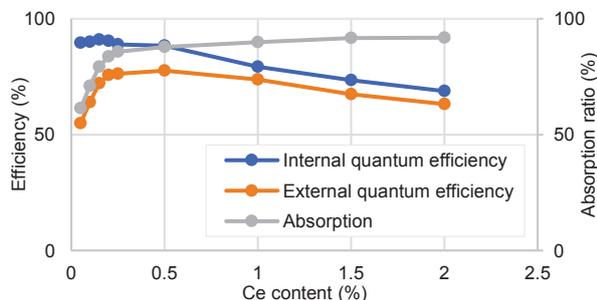


Fig. 5 Internal quantum efficiency, external quantum efficiency and absorption ratio for EPOCH-Neo samples as function of Ce content.

Figure 4 provides luminescence spectra obtained from specimens having Ce concentrations of 0.3mol%, 0.5mol% and 1.0mol%. The peak wavelength in the spectrum is seen to shift to longer wavelength with increasing Ce content.

Figure 5 plots the internal quantum efficiency, external quantum efficiency and absorption ratio for EPOCH-Neo specimens as a function of the Ce content, ranging from 0.05mol% to 2mol%. The internal quantum efficiency remained almost constant up to 0.5mol% and then decreased gradually. However, the absorption ratio increased rapidly up to 0.25mol% but the rate of increase was much slower thereafter. Thus, the external efficiency peaked at 0.5mol% Ce. In the case of the pure Ce:YAG, the optimal Ce concentration was higher than that for EPOCH-Neo and this variation is attributed to differences in the methods used to produce the two materials.

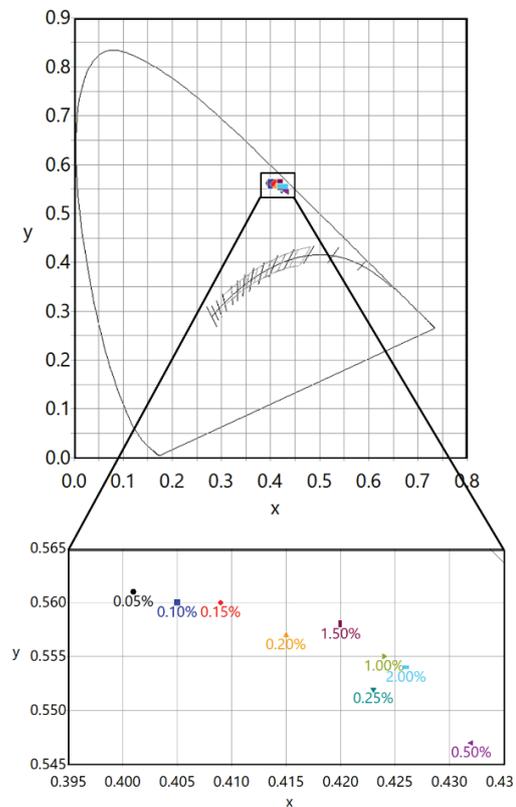


Fig. 6 Chromaticity coordinates for EPOCH-Neo specimens with Ce content ranging from 0.05mol% to 2.0mol%.

Figure 6 summarizes the chromaticity coordinates for EPOCH-Neo samples with Ce content ranging from 0.05mol% to 2.0mol%. Up to 0.5mol%, the coordinates moved toward the red region with increasing Ce content. However, above 0.5mol%, the coordinates were unchanged, indicating that a Ce content above 0.5mol% exceeded the solid solution range.

### 3. EPOCH-Neo Devices

Devices were manufactured by soldering  $5 \times 5$  mm EPOCH-Neo layers with varying thicknesses on  $17 \times 17 \times 2$  mm Cu-based heat sinks (Fig. 7).

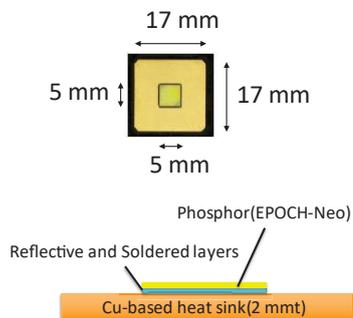


Fig. 7 Photographic image of EPOCH-Neo device and cross-sectional diagram.

Devices in which the Ce content was 0.5mol% and the thicknesses of the EPOCH-Neo layers were 0.1, 0.15, 0.2 and 0.25 mm were evaluated. Each EPOCH-Neo layer was coated with several metal layers prior to being soldered to the heat sink. These coatings functioned as a reflective layer, a corrosion protection layer and a thermal expansion buffer layer. Upon being exposed to high-power light, the temperature of the EPOCH-Neo layer increased and thermal expansion occurred. Consequently, there was the possibility of a rupture between the EPOCH-Neo layer and the heat sink due to the different thermal expansion coefficients for these two materials. Therefore, the device required a buffer. The reflective layer was made of Ag or Al and a Sn-Au alloy was used for soldering. The soldering process could have potentially corroded the reflective layer and so an anti-corrosion layer was included. The total thickness of all the layers was designed to be small enough to not obstruct the heat flow path.

### 4. Excitation with blue laser light

The EPOCH-Neo devices were excited by blue laser light and the resulting luminescence spectra were acquired using an Ocean Optics QEPro spectrometer (Fig. 8).

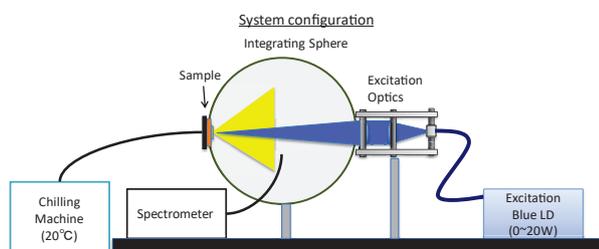


Fig. 8 Diagram of device characterization system.

The power of the blue laser was increased in

increments of 0.05 W from 0.5 to 19.98 W, and the laser beam was focused on the phosphor with a diameter of  $800 \mu\text{m}$ . The maximum excitation density was  $3.82 \text{ kW/cm}^2$ . During these trials, the temperature of the sample holder connected to the heat sink of the device was maintained at  $20 \text{ }^\circ\text{C}$  using a chiller. The resulting spectra were integrated between 465 and 780 nm and the luminous flux (in units of lm) was calculated. Figure 9 shows the relationship between the excitation power and the luminous flux for a device having an Ag reflective layer and a phosphor thickness of 0.2 mm. The luminous efficacy for each unit was calculated by dividing the luminous flux by the excitation power [W]. The luminous flux was found to steadily increase (Fig. 9) while the luminous efficacy [ $\text{lm/W}$ ] gradually decreased with increasing laser power from 2.31 to 19.98 W (Fig. 10). The luminous efficacy at 19.98 W was 92% of the original value. Other samples were characterized in the same manner. Table 1 summarizes the luminous efficacy values for devices having different phosphor thicknesses and different reflective layers. Following these trials, the appearance of each sample was examined and the optical properties were again evaluated. No damage was observed and the luminous characteristics of the units were not deteriorated (Table 1). These results demonstrated that the EPOCH-Neo devices were highly stable.

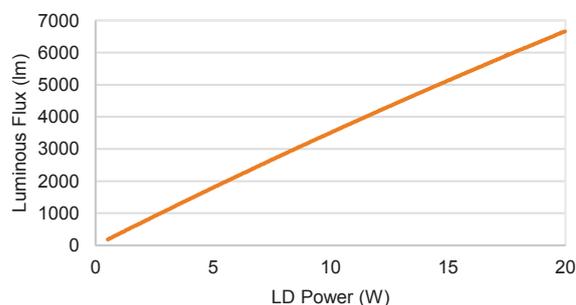


Fig. 9 Luminous flux for device with Ag reflective layer and phosphor thickness of 0.2 mm as function of excitation power.

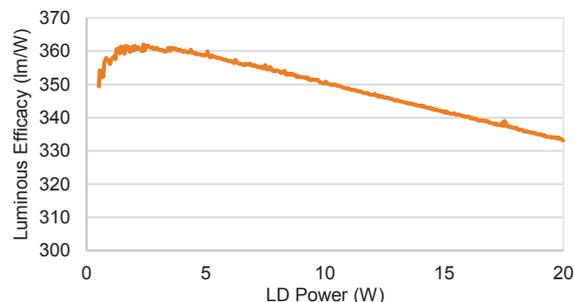


Fig. 10 Luminous efficacy of device with Ag reflective layer and phosphor thickness of 0.2 mm as function of excitation power.

Table 1 Proportion of luminous efficacy maintained by each specimen after testing.

		Phosphor thickness (mm)			
		0.1	0.15	0.2	0.25
Reflective layer material	Ag	91.2%	91.5%	92.0%	92.4%
	Al	91.1%	91.6%	91.9%	91.3%

### 5. Effects of reflective coating and phosphor thickness

Figure 11 shows the relationship between the phosphor thickness and the luminous flux of the devices when excited at a laser power of 19.98 W. These data were obtained for samples having Ag or Al reflective layers, and it is evident that the devices with the Ag layer provided higher luminance. The luminous flux increased up to a thickness of 0.15 mm but plateaued after this point.

Figure 12 plots the chromaticity coordinates for devices with different phosphor thicknesses and Ag reflective layers in response to excitation at 19.98 W. When the layer thickness was increased, the intensity of the reflected blue LD light decreased while the phosphor emission intensity increased. The emission from the phosphor was shifted to longer wavelength with increasing phosphor thickness as a consequence of self-absorption. Thus, the values of the x and y coordinates increased with increasing phosphor thickness. As a result, the color of the luminescence changed to yellow with increasing thickness.

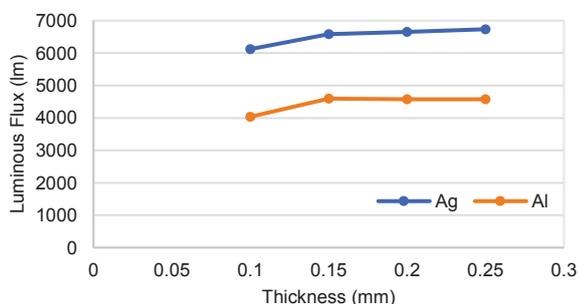


Fig. 11 Luminous flux as function of phosphor thickness for different reflective layer materials.

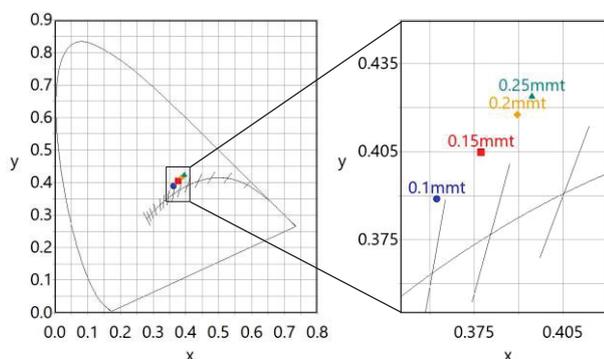


Fig. 12 Chromaticity diagram for specimens with Ag reflective layers.

### 6. Conclusions

The newly developed EPOCH-Neo phosphor, comprising a eutectic mixture of Ce:YAG and sapphire, was evaluated. Optical properties such as temperature quenching and luminous efficacy were ascertained. Thermal quenching for EPOCH-Neo was much less than that for commercial Ce:YAG powder at 275 °C. The absorption ratio, internal quantum efficiency and external quantum efficiency were determined for specimens with various Ce contents. The external quantum efficiency had exhibited a maximum at a Ce content of 0.5mol%. The chromaticity coordinates for EPOCH-Neo samples with Ce contents ranging from 0.05mol% to 2.0mol% were found to shift towards the red region as the Ce content was varied from 0.05mol% to 0.5mol%. Above 0.5mol%, the coordinates were constant, suggesting that 0.5mol% is the optimum Ce content.

Phosphor devices consisting of EPOCH-Neo, several different functional layers and a heat sink were evaluated under high-power blue laser irradiation. The laser power was changed from 0.5 to 19.98 W with a beam diameter of 800 μm and the devices were found to maintain over 90% of their original luminous efficacy. No damage was observed following these experiments. The chromaticity coordinates were found to move towards yellow with increasing thickness of the EPOCH-Neo layer, indicating a means of controlling these coordinates. This material appears to function as a suitable phosphor for use with strong light sources when excited by a high-power blue laser.

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### Reference

- [1] H.Morita, Y.Maeda, I.Kobayashi, Y.Sato, T.Nomura and H.Kikuchi, "Highly Efficient Laser-Phosphor Light Source for Projector," IDW 2017 p.620 (2017)
- [2] Masafumi Yoshimura, Shin-ich Sakata, Hisayoshi Iba, Takafumi Kawano, Keigo Hoshikawa, "Vertical Bridgman growth of Al<sub>2</sub>O<sub>3</sub>/YAG:Ce melt growth composite", Journal of Crystal Growth 416 (2015) 100-105.
- [3] Masafumi Yoshimura, Shin-ich Sakata, Seiya Yamada, Toshinori Taishi, Keigo Hoshikawa, "The growth of Al<sub>2</sub>O<sub>3</sub>/YAG:Ce melt growth composite by the vertical Bridgman technique using an a-axis Al<sub>2</sub>O<sub>3</sub> seed", Journal of Crystal Growth 427 (2015) 16-20.