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Preparation and Optical Properties of Transparent Ce:YAG Ceramics for High Power White LED

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Abstract. Transparent Ce:YAG ceramics were synthesized from the Ce:YAG powder which was produced by co-preparation method of the hydroxides. The Ce:YAG ceramics exhibit a broad emission band peaked at 530 nm due to the $5d \rightarrow 4f$ transition of Ce^{3+} . The transmittances of the samples obtained were 70 ~ 87 % at 800 nm. The absorption coefficient and emission intensity of Ce^{3+} were increased with the increase of the thickness. With increasing thickness of the sample, the color coordinates of the Ce:YAG ceramics under 465 nm LED excitation shifted from the blue region to the yellow region with passing nearby the theoretical white point. The highest value of luminous efficacy of the white LED was 73.5 lm/W.

1. Introduction

White LEDs are now extensively spotlighted as general illumination as well as backlight of the liquid crystal display (LCD), car headlights and indicator light. The advantages of the LEDs are low electric consumption, high brightness and long lifetime [1]. The most common method to make the white LEDs is combining of a blue LED and $Ce:Y_3Al_5O_{12}$ (Yttrium Aluminum Garnet) (Ce:YAG) phosphor packed with organic resins [2]. The function of the Ce:YAG phosphor is to absorb the blue light emitted from the blue LED and converts it to yellow light. The phosphor has a broad band emission due to the $5d \rightarrow 4f$ transition of Ce^{3+} . The combination of two lights of the transmitted blue light and the fluorescent yellow light makes white light. However, this white LED has a disadvantage of thermal degradation of organic resins, which is caused by heat generation of LED chip. As a result, it catches up with the degradation of luminous intensity and the change of emission color.

In order to solve this problem, a new type white LED using a glass ceramic phosphor has been studied [3-5]. The merits of this inorganic material are excellent heat-resistance compared with polymers, availability of production process and ease of formability. For the merits mentioned above, this approach using inorganic materials is useful for the white LED. To further enhance the performance of the white LEDs, it is important to develop new inorganic materials that have superior optical property than that of glass ceramic phosphor.

We have interest in transparent polycrystalline ceramics that have been studied as laser and scintillator materials [6]. They have attracted a great attention because the optical property and the efficiency are comparable or superior to those of single crystals. They have advantages of high homogeneity, optical transparency and thermal conductivity.

The transparent polycrystalline ceramics have been fabricated by solid state reaction using the powder of the same chemical composition. To achieve transparent polycrystalline ceramics of high quality, a fine powder with no or little agglomeration is necessary. Typically, wet chemical methods have been used for powder synthesis. The co-precipitation method, which is one of the wet chemical methods, is possible to produce a fine powder of multi cation oxides with large scale [7].

In this study, transparent polycrystalline ceramics of Ce:YAG are reported. As noted above, the Ce:YAG phosphor is suitable for white LEDs. Besides, since the unit cell of the YAG crystal is a cubic structure, it is easy to fabricate the transparent polycrystalline ceramics of the YAG crystal with high transparency in comparison with that of some crystals having other crystal symmetry. The co-precipitation method was used for synthesis of the YAG powder. We also measured the optical properties such as optical transmittance, photon distribution spectrum of total flux, color coordinates, luminous efficacy and luminous intensity under blue LED excitation.

2. Experimental

2.1. Synthesis of Ce:YAG ceramics

$\text{Y}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$, $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ and $\text{Ce}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ were used as the starting materials. They were dissolved in distilled water with a stoichiometric ratio for Y:Al:Ce of (2.997:5:0.003). The total ion concentration was adjusted at about 0.5 mol/l.

In the co-precipitation method, the precursor precipitate was produced by adding a NH_4HCO_3 solution of 2.5 mol/l at a speed of 10 ml/min in a beaker under mild stirring at 35 °C. The pH value was kept at 6.2. It was stirred to react completely for 72 h. The resulting precipitate was centrifuged and washed seven times with distilled water and ethanol. Then, it was oven-dried at 80 °C for 12 h and at 200 °C for 72 h. The precursor powders obtained were heated at 800 °C for 100 h in air to remove the organic substances. The powders were heated at 1200 °C for 90 min in air to get the YAG powder.

The milling process was carried out to grind a wide variety of the powder size into 200 ~ 300 nm averages. In this process, ethanol, a dispersant (Kyoeisha Chemical Co., LTD, Flowlen G-700) and SiO_2 were added to the powder at the weight ratio for powder:ethanol of (1:5), 0.4 wt% of the ethanol and 0.2 wt% of the powder, respectively.

After the milling process, a binder (Sekisui Chemical Co., LTD, S-LEC BL-1) was added in the slurry at 0.5 wt% of the powder and the slurry was milled with nylon balls for 24 h. Then, the slurry was poured into a mold located on a gypsum plate. It was dried at room temperature for 72 h. The caked slurry was heated to remove the organic substances at 800 °C for 100 h under O_2 atmosphere. Finally, it was heated at 1780 °C for 20 h under vacuum. The YAG ceramics obtained were cut and polished carefully. The thickness was set in the range of 0.1 ~ 0.8 mm.

2.2. Measurement

The microstructure of the YAG ceramics was observed by a confocal scanning laser microscopy (Olympus Corp., OLS1200). The optical transmittance of the mirror-polished samples on both surfaces was measured over the wavelength region from 200 to 850 nm using a spectrophotometer (Shimadzu Corp., UV-3600).

The photoluminescence (PL) of the Ce:YAG ceramics was measured under 465 nm LED (Nichia Corp., NFSB036B). In the optical measurement, the Ce:YAG ceramics were mounted on the LED. The spectral power and photon distribution of total radiant flux of PL spectra were measured by using an integrating sphere (Labsphere, Inc., LMS-100) which was connected to a CCD detector (Ocean Optics, Inc., USB2000) with an optical fiber. A standard halogen lamp (Labsphere, Inc., CLS-600) was used for calibrating this measurement system. The luminous efficacy (lm/W) of the Ce:YAG ceramics was calculated from the spectral power distribution and the electric power of the LED. The luminous intensity and color coordinates (x, y) of the Ce:YAG ceramics were measured from the calibrated spectra.

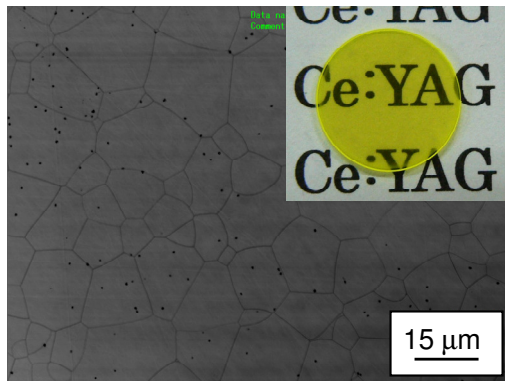


Figure 1. Confocal scanning laser micrograph of Ce:YAG ceramics. (Inset) Appearance of the mirror-polished specimen.

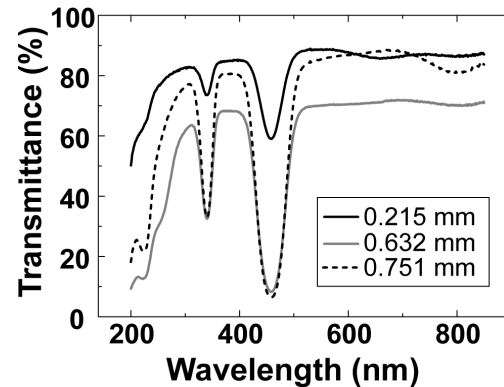


Figure 2. Optical transmittance of Ce:YAG ceramics.

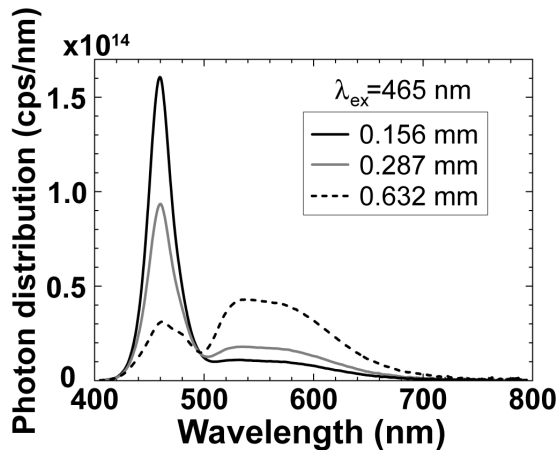


Figure 3. Photon distribution spectra of Ce:YAG ceramics under 465 nm LED excitation.

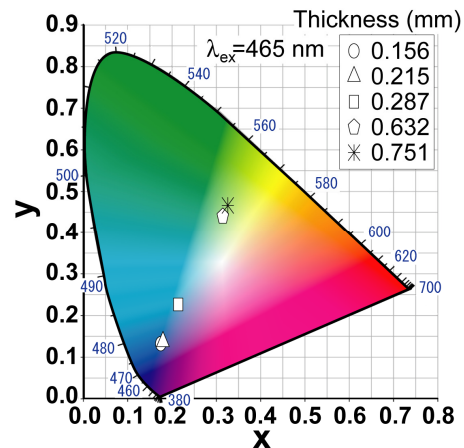


Figure 4. Chromaticity color coordinates of Ce:YAG ceramics under 465 nm LED excitation.

3. Results and Discussions

The confocal scanning laser micrograph of the Ce:YAG ceramics is shown in figure 1. The sample consisted of grains of several tens of micrometers, and pores were entrapped in the grains. The appearance of the mirror-polished specimen is shown in inset of figure 1. The pallet was 10.5 mm in diameter and 0.632 mm in thickness. It can be seen that the sample has good transparency.

The optical transmittance of the Ce:YAG ceramics is shown in figure 2. The transmittances of all the samples were 70 ~ 87 % at 800 nm. The absorptions at 330 nm and 460 nm were proportional to the thickness. These absorptions originate from the $5d \leftarrow 4f$ transition of Ce^{3+} . The transmittances of all the samples were flat against the measured wavelength region.

The photon distribution spectra of the PL of the Ce:YAG ceramics under 465 nm LED excitation is shown in figure 3. Broad emission band peaked at 530 nm originates from $5d \rightarrow 4f$ transition of Ce^{3+} . The intensity ratio of excitation and emission light was varied with the sample thickness. The PL intensity of 0.632 mm-thick sample was larger than the transmitted excitation light at 465 nm.

The chromaticity color coordinates of the Ce:YAG ceramics under 465 nm LED excitation are plotted on the CIE-1931 chromaticity diagram [8] in figure 4.

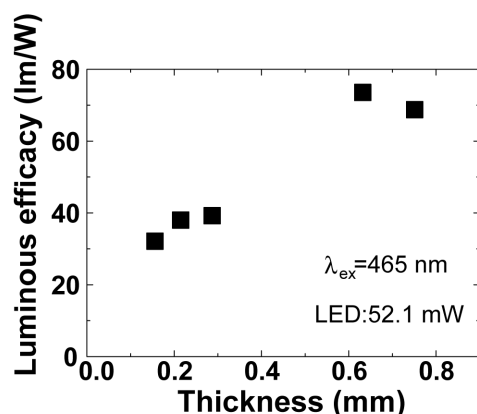


Figure 5. Thickness dependence of luminous efficacy of Ce:YAG ceramics under 465 nm LED excitation. Power dissipation of the LED was 52.1 mW at 20 mA.

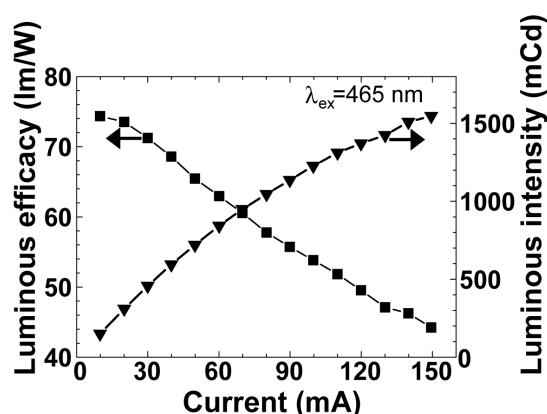


Figure 6. LED's current dependence of luminous efficacy and luminous intensity of Ce:YAG ceramics under 465 nm LED excitation (sample thickness: 0.632 mm).

The color coordinates of the combination of excitation and emission light shifted greatly from blue to yellow region with increasing thickness of the sample. The color coordinates of 0.632 mm-thick sample were located near the theoretical white point of (0.33, 0.33). This result shows that the white color can almost be achieved in the Ce:YAG ceramics under 465 nm LED excitation by controlling the sample thickness.

The thickness dependence of luminous efficacy of the Ce:YAG ceramics under 465 nm LED excitation is shown in figure 5. The power dissipation of the LED was 52.1 mW at 20 mA. The value of the luminous efficacy was in the range of 32.1 ~ 73.5 lm/W and increased with the increase of the thickness. The highest value of the luminous efficacy in the 0.632 mm-thick sample was 73.5 lm/W which is comparable to that of commercially produced white LED.

The LED's current dependence of luminous efficacy and luminous intensity for the 0.632 mm-thick sample under 465 nm LED excitation is shown in figure 6. The luminous efficacy was decreased from 74 lm/W to 44 lm/W with the increase of the current. The luminous intensity was increased from 100 mCd to 1550 mCd with the increase of the current. The decrease of the luminous efficacy was caused by the degradation of the luminous efficacy of the blue LED used with the increase of the current.

4. Conclusions

Ce³⁺-doped YAG ceramics were prepared by the vacuum sintering with the Ce:YAG powder which was produced by co-precipitation method using the hydroxides. Although pores were slightly entrapped in the grains, the sample had good transparency. The absorption coefficient and emission intensity of Ce³⁺ was increased proportionally to the sample thickness. The emission color of the combination of the blue (LED) and the yellow (Ce:YAG ceramics) lights can be changed by controlling the thickness. The highest value of the luminous efficacy was 73.5 lm/W, which is comparable to that of commercial white LEDs. The Ce:YAG ceramics can be a promising candidate phosphor for white LEDs.

Acknowledgments

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