Dyes and Pigments 139 (2017) 688–692

Contents lists available at ScienceDirect

Dyes and Pigments

journal homepage: www.elsevier.com/locate/dyepig

Design of laser-driven high-efficiency Al_2O_3/YAG:Ce^{3+} ceramic converter for automotive lighting: Fabrication, luminous emittance, and tunable color space

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ARTICLE INFO

Article history:
Received 29 October 2016
Received in revised form
28 December 2016
Accepted 30 December 2016
Available online 3 January 2017

Keywords:
Laser diode
$Y_3Al_5O_{12}$: Ce^{3+}
Ceramic phosphor plate
Automotive lighting
Luminous flux

ABSTRACT

An $Al_2O_3/Y_3Al_5O_{12}:Ce^{3+}$ ceramic phosphor plate (CPP) for which a nano-phosphor is used for a high-power laser diode (LD) application for white automotive-lighting is reported here. The prepared CPP shows improved luminous properties that are owing to the light propagation of the hexagonal α-Al_2O_3 in the CPP. The amount of α-Al_2O_3 added to the $Y_3Al_5O_{12}:Ce^{3+}$ CPP was optimized, and its impact on the luminous characteristics was investigated. The luminous properties of the $Al_2O_3/Y_3Al_5O_{12}:Ce^{3+}$ CPP are improved when compared with the $Y_3Al_5O_{12}:Ce^{3+}$ CPP, and the luminous emittance, as well as the conversion efficiency, is therefore also improved. The results of the present study indicate that the $Al_2O_3/Y_3Al_5O_{12}:Ce^{3+}$ CPP can serve as a potential material for the solid-state laser lighting in automotive applications.

1. Introduction

Light-emitting diodes (LEDs) are the most widely used light sources for solid-state lighting (SSL) [1]. LEDs utilize the electric current to generate light. Energy is emitted as light when it is under a forward bias in the p-n junction, holes are injected from the *p*-type region, and electrons from the *n*-type region combine together in the depletion region, and light is consequently generated [2,3]. LEDs have been favoured in the solid-state-light industry due to their long life, high efficiency, and the absence of toxic materials, along with the flexibility of the designs when compared with the conventional lighting choices [4,5].

However, LEDs that undergo a saturation of the output power with an increasing forward-bias current leads to a deterioration of the luminous efficacy [6], which is called an "efficiency droop." Alternatively, laser diodes (LD) do not suffer from these problems and provide faster switching, a narrower emission spectrum, and a high intensity; therefore, LDs have recently come into the spotlight as the most-efficient high-power sources [7,8]. LDs are applicable to projections, medical-industry uses, and automotive lighting because of the advantages of directionality, a compact size, a high luminous flux, and a low power consumption [9]. The general approach for the generation of white light using blue LEDs and LDs involves the use of yellow-emitting phosphors (YAG: Ce^{3+}) [10]. The YAG: Ce^{3+} absorbs the blue light and emits a down-converted yellow light. The combination of the remaining blue component and the frequency down-converted yellow-centered wide spectrum is what is seen as the "white"-light output from a device such as an LED [11]. YAG: Ce^{3+} shows a broad emission spectrum in the range of 500 nm–700 nm, which is centered at 550 nm, and a strong absorption band in the blue region [12]. The quantum efficiency of YAG: Ce^{3+} is high (>85%), and it shows a stability under humid conditions [13]. To manufacture the white LED, the formation of the conventional structure involves the spreading and hardening of the phosphor in silicone (PiS) on a blue-LED chip;

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http://dx.doi.org/10.1016/j.dyepig.2016.12.071
0143-7208/

however, the mixture absorbs the heat emitted from the LED chip and cannot release it, thereby deteriorating the efficiency of the phosphor, and thermal quenching is consequently induced [14,15]. For this reason, the application of the PiS type to the LD system is inadequate.

To overcome the problems, the YAG: Ce^{3+} single crystal (SC) and polycrystalline ceramic phosphor plate (CPP) have garnered attention with respect to the LD system [16,17]. Recently, both the YAG: Ce^{3+} SC and CPP were identified as candidates with high thermal quenching, and mechanical and chemical properties were also reported [18]. In the case of the mentioned materials, the converted light is trapped due to the gap of a large refractive-index value (n) between the YAG: Ce^{3+} CPP ($n = 1.82$) and the air ($n = 1$). The trapped light emits the side of YAG: Ce^{3+} CPP, as well as performing a re-absorption, so that the light is converted to heat [19]. To the authors' best knowledge, these matters lead to the deterioration regarding white-color uniformity and quality called the “yellow-ring effect.”

Here, an attempt was made to add the hexagonal α-Al_2O_3 particle as the second phase in the cubic YAG: Ce^{3+} CPP for the improvement of both the yellow-ring phenomenon and the light-extraction efficiency. Generally, Al_2O_3 with a representative of transparent ceramic materials has a refractive index value ($n = 1.76$). Also, the α-Al_2O_3 particle has a birefringence effect due to its hexagonal structure, and its scattering of the light reduces the yellow-ring effect. Zhang et al. reported on the composite-phase ceramic-phosphor luminous efficacy of ~95 lm/W for the white-LED application when the mole ratio of α-Al_2O_3/YAG: Ce^{3+} is 0.65; however, a two-phase sintering of the YAG: Ce^{3+} CPP with α-Al_2O_3 for white-light generation in LD applications such as a display, lamp, and automotive-lighting sections has not been reported.

In this paper, the advanced research of the authors regarding the α-Al_2O_3/YAG: Ce^{3+} CPP, for which the nano-structured YAG: Ce^{3+} precursor is directly combined with blue LDs for white-light generation, is reported. The authors substitute the organic-resin mixture with phosphor powder for the YAG: Ce^{3+} CPP. The synthetic technique, sintering condition, and composition tuning of the Al_2O_3/YAG: Ce^{3+} phosphor are performed. The optical properties of the YAG: Ce^{3+} CPP are also optimized.

2. Experimental

The YAG: Ce^{3+} phosphors were synthesized according to the co-precipitate homogeneous method. All chemicals were used without further purifications. The aluminum nitrate nonahydrate (Sigma-Aldrich, ≥98% pure), ammonium aluminum sulfate dodecahydrate (Sigma-Aldrich, ≥98% pure) yttrium nitrate (Sigma-Aldrich, 99.8%), cerium nitrate (Sigma-Aldrich, 99%) and urea (Sigma-Aldrich, 99.5%) were purchased from Sigma-Aldrich. Firstly, yttrium nitrate and cerium nitrate were dissolved in de-ionized water for 0.1 M solution A. aluminum nitrate nonahydrate and ammonium aluminum sulfate dodecahydrate were dissolved in de-ionized water for 0.1 M solution B. For stoichiometric composition of YAG: Ce^{3+}, Solution A and B is mixed with ratio of 3:5 and then vigorously stirred for 30 min. After completely mixed, urea for 0.08 mol is added and stirred into mixed solution at 90 °C for 3 h. The resultant mixture was aged for 2 h 30 min at room temperature. The precipitate was isolated *via* centrifugation, washed with deionized water and ethanol several times, and then dried with a lyophilizer. The resulting powder was annealed at 1100–1200 °C for 4 h under a reducing nitrogen atmosphere containing 5% H_2 gas. A detailed synthesis procedure of YAG: Ce^{3+} is presented in Supplementary Information Fig. S1. The fabrication of the Al_2O_3/YAG: Ce^{3+} CPP was conducted with ball milling using the YAG: Ce^{3+} phosphor and SiO_2 as a sintering aid. All of the materials were milled with a ball-milling machine using Al_2O_3 balls with a diameter of 5 mm for 24 h. These mixtures were dried at 90° C for 12 h and then, through a uniaxial pressing under 20 MPa, they were fabricated into a disk with a diameter of 10 mm and a thickness of 5 mm. The disks were fired at 1000° C for 6 h under an air atmosphere to remove the organic materials, followed by an isostatic cold-pressing under 300 MPa for 30 min. The disks were then sintered with a graphite-heated vacuum furnace (10^{-3} Pa) at 1600° C for 12 h. The sintered YAG: Ce^{3+} CPP was annealed at 1450° C for 24 h under an air atmosphere to remove the oxygen vacancies. The Al_2O_3/YAG: Ce^{3+} were then treated using mirror polishing. A detailed fabrication of YAG: Ce^{3+} CPP is shown in Supplementary Information Fig. S2. The crystalline phase of the YAG: Ce^{3+}phosphor was identified using powder X-ray diffraction (XRD, D-MAX 2500, Rigaku, Tokyo, Japan) and a CuKα target with a $10° \leq \theta \leq 80°$ alignment. The surface morphology of the YAG: Ce^{3+} CPP was observed using field-emission SEM (FE-SEM, JSM-7600F, JEOL, Tokyo, Japan). The luminous properties of the prepared samples were measured with double integrating spheres (PSI Co., Ltd/Korea) under blue-laser excitation (445 nm blue LD) with the use of handmade LG Electronics equipment (Seoul, Korea).

3. Results and discussion

The XRD patterns and 450 nm laser-based photoluminescence (PL) results synthesized at 1200 °C are shown in Fig. 1. The synthesized YAG: Ce^{3+} CPP completely corresponds to the diffraction patterns of the pure YAG (JCPDS card no.33-0040) without other phases such as CeO_2, Y_2O_3, Al_2O_3, $YAlO_3$, or $Y_2Al_4O_9$, as shown in Fig. 1a. Fig. 1b depicts the laser-PL spectra of the YAG: Ce^{3+} CPP. The YAG: Ce^{3+} CPP indicates a high-efficiency PL intensity with an increase of the forward-bias current that is centered at 548 nm with a 445 nm blue-laser excitation, leading to a broadband emission at a wavelength. This emission is ascribed to the electron transitions from the lowest crystal-field splitting component of the 5d level to the ground state of the Ce^{3+} ion that is attributed to $^2F_{5/2}$ and $^2F_{7/2}$ [20]. With the increasing of the forward-bias current, the PL-emission intensity is improved because of the effective conversion from blue to yellow; however, the PL-emission intensity is decreased over the forward-bias current of 760 mA, and this is attributed to the thermal-quenching behavior at higher operating currents for the LDs, which implies that the brightness of the garnet-based CPP cannot be further improved.

To confirm the automotive-lighting applications, the YAG: Ce^{3+} CPP was analyzed with the 445 nm blue-emitting LD. For the application of the YAG: Ce^{3+} CPP in high-power LDs, it is essential to analyze the luminous characteristics of the YAG: Ce^{3+} CPP including the luminous emittance, conversion efficiency, surface analysis, and correlated color temperature (CCT) under the blue-laser excitation of 445 nm. Fig. 2a demonstrates the luminous illumination of the white-LD package using the garnet-based CPP under the blue-laser excitation of 445 nm. By increasing the blue-radiant flux, the luminous-illumination value of the garnet-based CPP linearly increased, which is ascribed to the increased number of electrons that have been elevated to the excited state of the Ce^{3+} ions. The luminous emittance was increased without saturation at up to 13.5 W/mm^2, and this is attributed to the effective conversion of the blue light to yellow for the white-light conversion. The highest luminous-illumination value of 1495 lm/mm^2 in the garnet-based CPP was obtained at the blue radiant-flux density of 13.5 W/mm^2 when the Ce^{3+} contents are at 0.011 mol, implying that the garnet-based CPP can be applied as a color converter for high-power white-laser light. With the increase of the Ce^{3+} concentration, the luminous emittance value continuously increased;