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Component Regulation and Performance Optimization of Al_2O_3-YAG:Ce Composite Ceramic Phosphors for High-power Laser Lighting

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Abstract: Solid-state laser lighting fabricated by combining blue laser diodes (LDs) and yellow-emitting phosphor converters has attracted great attention in high-luminance applications. However, the achievement of high-power laser lighting is significantly affected by the thermal quenching effect of phosphor converter materials. Therefore, component regulation and performance optimization are required to improve the thermal conductivity and luminescence uniformity of phosphor converters. In this work, a series of Al_2O_3-YAG:Ce composite ceramic phosphors with different Al_2O_3 contents were prepared by solid-state reaction sintering. The influences of Al_2O_3 contents on the microstructure, phase composition, optical properties and thermal performance of the Al_2O_3-YAG:Ce ceramic phosphors were investigated in detail. The total transmittance of the Al_2O_3-YAG:Ce ceramic phosphors at 800 nm tends to decline (82.6%→23.6%) with the increase of Al_2O_3 content (0→90%, weight ratio). Both excitation and emission intensity of the Al_2O_3-YAG:Ce ceramic phosphors initially increase and then decrease with increasing Al_2O_3 content. When the weight ratio of Al_2O_3/Al_2O_3-YAG:Ce is 70%, the ceramic phosphor exhibits a high thermal conductivity of 25.7 $W\cdot m^{-1}\cdot K^{-1}$ at room temperature and the highest emission intensity. A high luminous flux of 3724 lm and luminous efficacy of 239.4 $lm\cdot W^{-1}$ are obtained when pumping the 70% Al_2O_3-YAG:Ce ceramic phosphor with blue LDs at a power density of 20 $W\cdot mm^{-2}$. Additionally, the luminous efficacy only decreases by 10.5% and the luminous flux continues to increase without showing luminescence saturation, when the power density increases from 1 $W\cdot mm^{-2}$ to 20 $W\cdot mm^{-2}$. Therefore, the Al_2O_3-YAG:Ce composite ceramic phosphors are promising in high-power laser lighting for excellent luminous efficiency and improved thermal stability.

Key words: Al_2O_3-YAG:Ce; laser lighting; luminous efficacy; component regulation; performance optimization

In recent years, solid-state lighting (SSL) is gradually replacing traditional incandescent lamps owing to high efficiency, low pollution, and long service life[1-2]. The common way to obtain white light-emitting diodes (w-LEDs) is to combine blue light-emitting diode (LED) chips with yellow-emitting $Y_3Al_5O_{12}:Ce^{3+}$ phosphors encapsulated by organic binders[3-4].

The commercial w-LEDs are currently limited by efficiency droop, which can be interpreted as the saturation of the output power with increasing input current density, so it is difficult to achieve high-power and high-brightness lighting. However, laser diodes (LDs) can maintain high efficiency even at a high input laser power density of 25 $kW\cdot cm^{-2}$, and consequently allow for a high output luminous flux[5-8]. As a result, LD chips combined with phosphor converters are considered as a promising candidate for high-power lighting[9-10]. The superiority of LD chips over the LED chips leads to higher requirements for the phosphor converters. The excellent heat dissipation performance and thermal stability are required for the phosphor converters to realize high-power lighting under high power density laser

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irradiation and thermal attack.

Among many kinds of phosphors, YAG:Ce material possesses many excellent properties, such as strong absorption of blue light, high quantum efficiency (>85%), broadband emission, high chemical and thermal stability[11-13]. At present, the most widely used phosphor converter is manufactured by embedding YAG:Ce powders in organic binders[14], but the low thermal stability restricts its further applications in high-power laser lighting. To solve the problem, the Al_2O_3-YAG:Ce composite ceramic phosphors were designed by adding Al_2O_3 as the secondary phase to improve the thermal conductivity and light scattering of YAG:Ce ceramics[15-17]. Al_2O_3 has been chosen as the optimum secondary phase because of its unique characteristics. The thermal conductivity of Al_2O_3 (32–35 $W\cdot m^{-1}\cdot K^{-1}$) is much higher than that of YAG (9–14 $W\cdot m^{-1}\cdot K^{-1}$) at room temperature[18-20]. The thermal expansion coefficient of Al_2O_3 (8.4×10^{-6} K^{-1}) is close to that of YAG (8.0×10^{-6} K^{-1}), which means that Al_2O_3 can well match YAG:Ce[21-22]. In addition, when irradiated by high- power laser diodes, Al_2O_3 ceramics exhibit excellent chemical stability. Based on the above analyses, the Al_2O_3-YAG:Ce composite ceramic phosphors are considered as ideal composite ceramic phosphors for high-power laser lighting.

Recently, the Al_2O_3-YAG:Ce composite ceramic phosphors have been widely studied. Li, *et al*[23] designed the translucent Al_2O_3-YAG:Ce composite phosphor ceramics with high thermal conductivity of 18.5 $W\cdot m^{-1}\cdot K^{-1}$ and excellent thermal stability (only decreases by 10.5% at 200 ℃), which shows no luminescence saturation even under 50 $W\cdot mm^{-2}$ laser excitation. Liu, *et al*[24] prepared the Al_2O_3-YAG:Ce ceramic phosphor with a high luminous emittance of 1888 $lm\cdot mm^{-2}$ when combined with blue LDs. Xu, *et al*[25] found the secondary phase Al_2O_3 plays a vital role in the performance of the Al_2O_3-YAG:Ce composite ceramics for high-power laser lighting applications. With the increase of Al_2O_3 content, the Al_2O_3-YAG:Ce composite ceramics show alleviative luminescence saturation and enhanced reliability. Nevertheless, there is lack of evidence about the influence of different weight ratios of Al_2O_3/Al_2O_3-YAG:Ce on the microstructure and optical properties of Al_2O_3-YAG:Ce composite ceramic phosphors when combined with blue LDs in a reflection mode.

In this work, a series of Al_2O_3-YAG:Ce composite ceramic phosphors with different weight ratios of Al_2O_3 to xAl_2O_3-YAG:Ce (denoted as xAl_2O_3-YAG:Ce, where x is the weight ratio of Al_2O_3 to Al_2O_3-YAG:Ce) were fabricated by the solid-state reaction and vacuum sintering method. The synthetic technique and composition tuning of Al_2O_3-YAG:Ce composite ceramic phosphors were performed. The luminous properties such as luminous flux and luminous efficiency by combining the prepared ceramics with a high-power blue laser were measured. Moreover, the suitability of Al_2O_3-YAG:Ce used as phosphor converters for high-power laser lighting was evaluated.

1 Experimental

Commercial Y_2O_3 (99.999%, Fujian Changting Golden Dragon Co., Ltd), α-Al_2O_3 (99.99%, Fenghe Ceramic Co., Ltd), CeO_2 (99.995%, Fujian Changting Golden Dragon Co., Ltd) were used as raw materials. A series of ceramic phosphors (Al_2O_3-YAG:Ce) with different Al_2O_3 contents were fabricated by the solid-state reaction method. The Ce concentration was fixed at 0.3% (in atomic fraction), and the weight ratio of Al_2O_3/Al_2O_3-YAG:Ce was in the range of 0–90%. 0.5% tetraethyl orthosilicate was used as the sintering additive. The raw powders were weighed according to the stoichiometric ratio and mixed for 12 h by a planetary ball mill, with ethanol as a dispersant and alumina balls as the grinding media. After being dried at 70 ℃ for 2 h and sieved through a 200-mesh screen (ϕ74 μm), the powder mixtures were calcined at 600 ℃ in air for 4 h to remove the organic impurities. Then they were pressed into disks with a diameter of 20 mm under 20 MPa and cold isostatically pressed under 250 MPa for 3 min. The pretreated disks were sintered at 1750 ℃ under the vacuum of 4.0×10^{-4} Pa for 10 h and annealed at 1450 ℃ in air for 10 h. Eventually, the composite ceramic phosphors were double-surface polished to 1 mm for further measurements. Sintered samples were thermally etched at 1450 ℃ for 3 h before SEM measurements.

X-ray diffraction (XRD) patterns of these composite ceramic phosphors were obtained using an X-ray diffractometer (XRD, Ultima IV, Rigaku, Japan), with CuKα radiation in the angle range of 2θ=15°–75°. The microstructure was characterized by a high-resolution field-emission scanning electron microscope (FESEM SU8220, Hitachi, Japan). The total transmittance of the ceramics over the wavelength range from 200 to 800 nm was tested by a UV-Vis spectrophotometer (Cary 5000, Varian Medical System Inc. Palo Alto, USA). The homemade multi-function combined fluorescence spectrum test system (SicOmni-I) was used for the UV-vis fluorescence test. The system was equipped with VX-XBO 150 W Xenon lamp, Omni-λ3007 (Zolix, China) monochromator and PMTH-S1-CR131 photomultiplier tube. The spectral wavelength was corrected by the LHM254 mercury lamp. The thermal conductivity was measured by a laser thermal conductivity analyzer

(LFA447, Netzsch, Germany). A reflection mode was applied to measure the luminous properties of the Al_2O_3-YAG:Ce composite ceramic phosphors under 450 nm laser excitation. The ceramic plates were glued to a heat sink and tested by an integrating sphere connected to a CCD spectrometer (OHSP-350, HOPOOCOLOR, China).

2 Results and discussion

The photograph of the Al_2O_3-YAG:Ce composite ceramic phosphors with different Al_2O_3 contents (0–90%) is shown in Fig. 1. The diameter and the thickness of the obtained ceramics are 15 and 1 mm, respectively. All xAl_2O_3-YAG:Ce ceramic phosphors exhibit bright yellow color, due to the strong absorption in the blue light range and high emission in the yellow light range. When the Al_2O_3 content is 0, the YAG:Ce ceramic phosphor shows high optical transmittance, while the xAl_2O_3-YAG:Ce composite ceramic phosphors turn opaque, since the Al_2O_3 particles can act as optical scattering centers, leading to the decrease of the optical transmittance.

XRD patterns of the composite ceramic phosphors with different Al_2O_3 contents are shown in Fig. 2. All diffraction peaks of YAG:Ce ceramic sample (x=0) can be indexed to YAG (JCPDS#33-0040), indicating that single phase YAG is successfully synthesized by the solid-state reaction method. Besides the diffraction peaks corresponding to YAG, the characteristic peaks of α-Al_2O_3 appear in the XRD patterns of the ceramic phosphors. The intensity of the Al_2O_3 diffraction peaks increases with the weight ratio of Al_2O_3/Al_2O_3-YAG:Ce increasing.

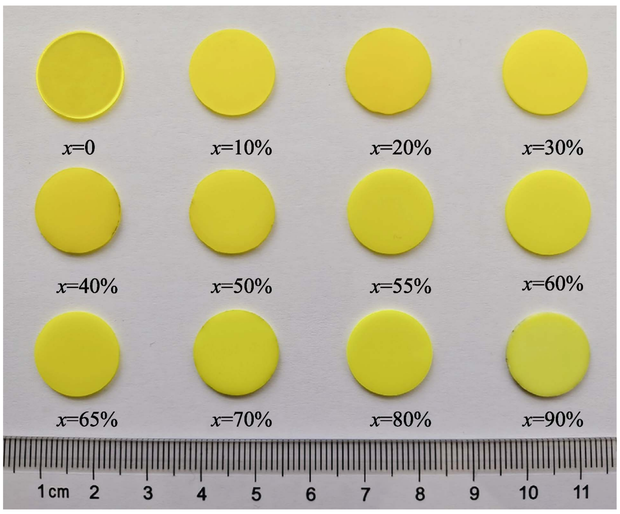

Fig. 1 Photograph of xAl_2O_3-YAG:Ce ceramic phosphors with thickness of 1 mm

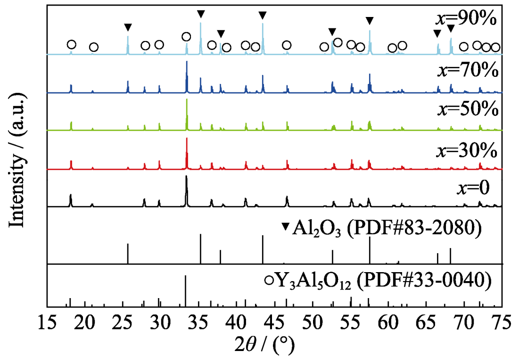

Fig. 2 XRD patterns of xAl_2O_3-YAG:Ce ceramic phosphors

SEM images of the thermally etched surfaces of Al_2O_3-YAG:Ce ceramic phosphors were carried out to investigate the link between the microstructure and Al_2O_3 content, as displayed in Fig. 3. Based on the imaging principle of SEM, bright and dark grains can be identified as YAG and Al_2O_3, respectively. The Al_2O_3 particles are embedded in the YAG matrix, and the dark area expands with the increase of the weight ratio of Al_2O_3 to Al_2O_3-YAG:Ce, leading to the decrease of the optical transmittance. Meanwhile, the grain size and distribution of Al_2O_3 and YAG:Ce in the composite ceramics are inhomogeneous, which may be an important factor affecting the absorption efficiency of blue light and the extraction efficiency of yellow light. Additionally, the high thermal conductivity of Al_2O_3 can provide a pathway to dissipate heat from the YAG matrix, which can be attributed to the interesting microstructure.

To evaluate the influence of different Al_2O_3 contents on the light scattering of the composite ceramic phosphors, the total transmittance of the ceramics with the thickness of 1 mm is characterized in Fig. 4(a). A wide absorption band appears in the 400–500 nm range, which is attributed to the 4f-5d$_1$ transition of Ce^{3+}. It is found that

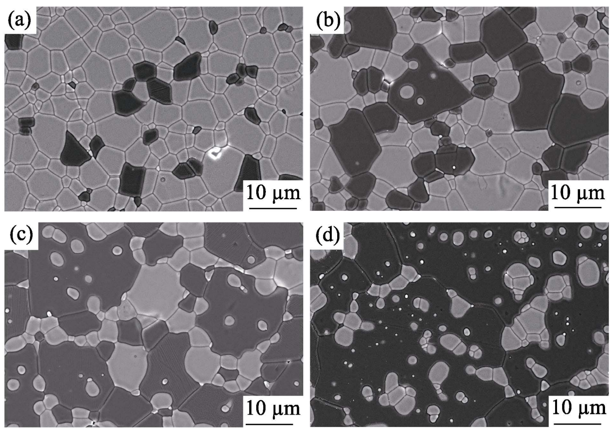

Fig. 3 SEM images of xAl_2O_3-YAG:Ce ceramic phosphors
(a) x=20%, (b) x=40%, (c) x=60%, and (d) x=70% thermally etched at 1450 ℃ for 3 h in air

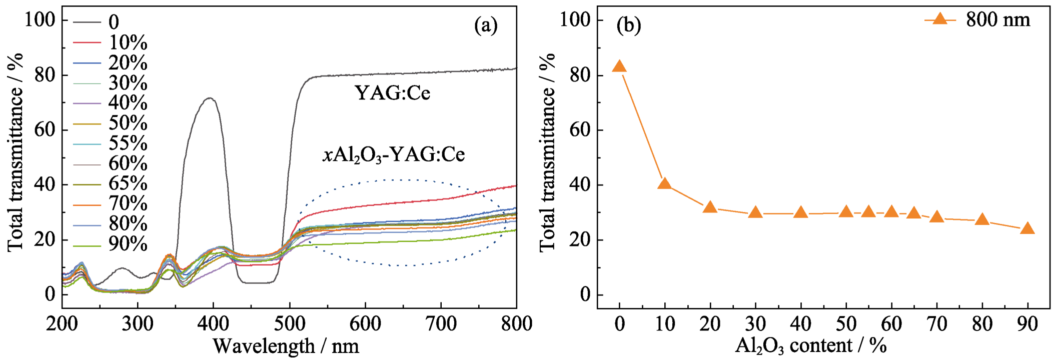

Fig. 4　Total transmittance of xAl$_2$O$_3$-YAG:Ce ceramic phosphors

(a) Total transmittance curves; (b) Total transmittance at 800 nm

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the total transmittance at 800 nm tends to decline (82.6%→23.6%) with the increase of Al_2O_3 content (0→90%), as shown in Fig. 4(b). Therefore, composite ceramic phosphors with appropriate transmittance can achieve optimal luminous efficacy after combining with blue LDs.

Fig. 5 shows the photoluminescence (PL) and photoluminescence excitation (PLE) spectra of the Al_2O_3-YAG:Ce composite ceramic phosphors. As seen in Fig. 5(a), no evident changes in the shape and position of the peaks can be observed with the increase of Al_2O_3 content. The PLE spectrum is characterized by two wide bands centered at 342 and 465 nm, attributed to the Ce^{3+} excitation transitions of 4f-5d$_2$ and 4f-5d$_1$, respectively[26]. The emission spectrum shows a broad band ranging from 510 to 640 nm. As a result, the addition of Al_2O_3 does not affect the luminescence performance of YAG:Ce because of the invariant local electronic environment around Ce^{3+}. As shown in Fig. 5(b), both excitation and emission intensity of the Al_2O_3-YAG:Ce composite ceramic phosphors initially increase and then decrease with Al_2O_3 content increasing. The 70% Al_2O_3-YAG:Ce ceramic phosphor exhibits the highest excitation and emission intensity. Therefore, it can be noticed that with the increase of Al_2O_3 content in the Al_2O_3-YAG:Ce composite ceramic phosphors, the concentration quenching effect is alleviated by reducing the concentration of Ce^{3+} ions, so that more blue light is absorbed and converted by Ce^{3+} to increase the luminescence.

To study the performance of the composite ceramic phosphors with different Al_2O_3 contents in the application of high-power laser lighting, the luminous properties of sintered ceramics were measured in a reflection mode where the ceramic phosphor was glued to an aluminum heat sink. According to the diagram of light propagation in Fig. 6(a), the Al_2O_3 particles make the direction of the light more variable, which can increase the absorption and conversion efficiency. As exhibited in Fig. 6(b), the luminous efficacy exhibits a monotonous enhancement as the Al_2O_3 content increases from 0 to 70% and a sharp decrease as the Al_2O_3 content exceeds 70%, which is consistent with the trend observed in Fig. 5. A maximum luminous efficacy of 267.4 $lm\cdot W^{-1}$ is obtained when pumping the 70% Al_2O_3-YAG:Ce ceramic phosphor with a blue laser at a power density of 1 $W\cdot mm^{-2}$, which is enhanced by 28.9% as compared to YAG:Ce ceramics. Luminous efficacy and luminous flux are plotted as a function of the blue laser power density (Fig. 6(c)). By increasing the power density, the luminous flux of the 70% Al_2O_3-YAG:Ce ceramic phosphor linearly increases, which can be attributed to the enhanced number of electrons elevated to the excited state of Ce^{3+}. A high luminous flux (3724 lm) can be

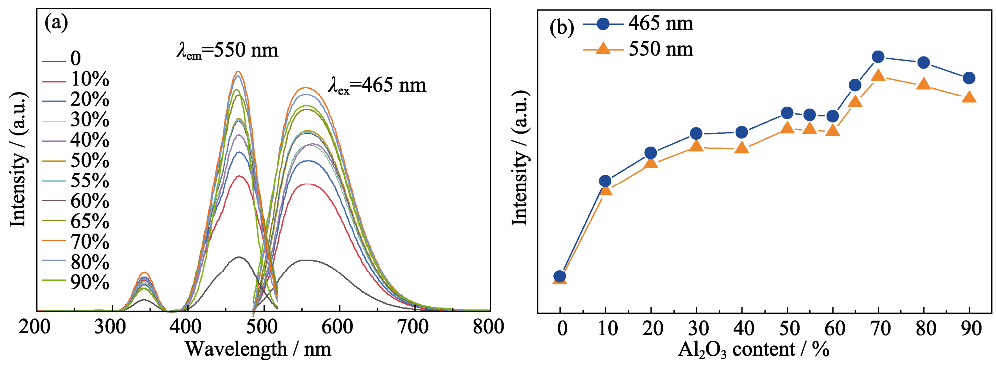

Fig. 5　PL and PLE spectra of xAl$_2$O$_3$-YAG:Ce ceramic phosphors

(a) PL and PLE spectra; (b) PL and PLE spectra intensity at 465 and 550 nm

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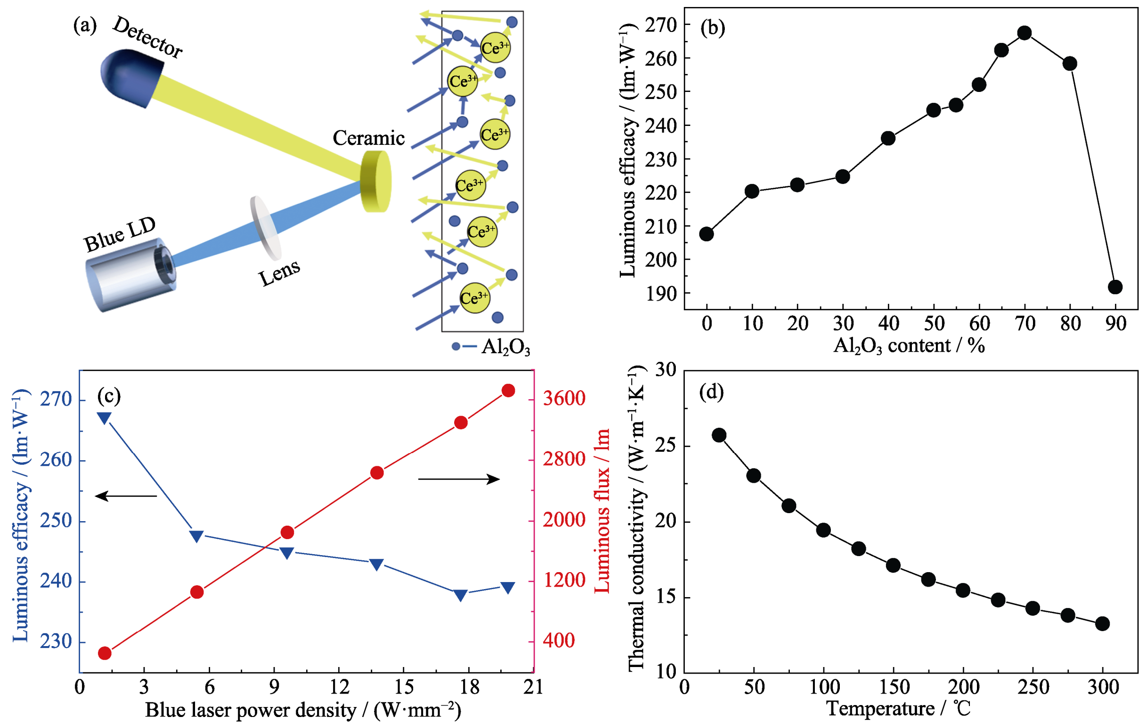

Fig. 6 Light propagation path, luminous efficacy, luminous flux and thermal conductivity of xAl_2O_3-YAG:Ce ceramic phosphors
(a) Diagram of reflection mode to measure luminous properties and light propagation; (b) Luminous efficacy of the ceramics with different Al_2O_3 contents under 1 W·mm^{-2} blue laser irradiation; (c) Luminous efficacy and luminous flux of 70% Al_2O_3-YAG:Ce as a function of blue laser power density; (d) Thermal conductivity of 70% Al_2O_3-YAG:Ce as a function of temperature from 25 ℃ to 300 ℃

obtained when the power density reaches 20 W·mm^{-2} (the upper limit of the laser device). By contrast, the luminous efficacy decreases by 10.5% when the power density increases from 1 to 20 W·mm^{-2} owing to the thermal quenching effect. The thermal conductivity is a vital parameter for high-power LD applications, which affects the service life and efficiency of the laser device. The thermal conductivity of the 70% Al_2O_3-YAG:Ce ceramic phosphor as a function of the temperature from 25 ℃ to 300 ℃ was characterized. As shown in Fig. 6(d), the thermal conductivity drops in response to the rise of the temperature. The thermal conductivity is 25.7 W·m^{-1}·K^{-1} at room temperature for the 70% Al_2O_3-YAG:Ce, which is considerably higher than YAG:Ce ceramics (9.6 W·m^{-1}·K^{-1})[27-28]. Due to the higher thermal conductivity, the composite ceramic phosphors can quickly dissipate the accumulated heat, which keeps the phosphor converter at a lower temperature to minimize the thermal quenching effect[29-30]. The above analysis implies that the Al_2O_3-YAG:Ce composite ceramic phosphors are promising in high-power laser lighting for excellent luminous efficiency and improved thermal stability.

3 Conclusion

In summary, a series of Al_2O_3-YAG:Ce composite ceramic phosphors with different weight ratios of Al_2O_3 to Al_2O_3-YAG:Ce were prepared by the solid-state reaction method in vacuum. The appropriate amount of Al_2O_3 particles distributed in the YAG:Ce matrix can not only improve the absorption and extraction efficiency of the light, but also improve the luminous efficacy and reliability. The microstructure, phase composition and optical properties of the ceramic phosphors with various Al_2O_3 contents were investigated. The 70% Al_2O_3-YAG:Ce ceramic phosphor exhibits a high thermal conductivity of 25.7 W·m^{-1}·K^{-1} at room temperature and the highest emission intensity. A high luminous flux of 3724 lm and luminous efficacy of 239.4 lm·W^{-1} are obtained when pumping the 70% Al_2O_3-YAG:Ce ceramic phosphor with blue LDs at a power density of 20 W·mm^{-2}. The results indicate that the designed ceramic phosphors can be used as a promising phosphor converter for high-power laser lighting.

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高功率激光照明用 Al_2O_3-YAG:Ce 复相陶瓷荧光体的组分设计与性能优化

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摘 要: 结合蓝色激光二极管和黄色荧光转换器制备的固态激光照明引起了人们极大的关注, 但荧光转换材料的热猝灭效应显著影响了高功率激光照明的实现。通过组分设计和性能优化可以提高荧光转换器的热导率和发光均匀性。本工作采用固相反应烧结技术制备了一系列不同 Al_2O_3 含量的 Al_2O_3-YAG:Ce 复相陶瓷荧光体, 研究了 Al_2O_3 含量对 Al_2O_3-YAG:Ce 陶瓷荧光体微观结构、相组成、光学性能和热学性能的影响。Al_2O_3-YAG:Ce 陶瓷荧光体在 800 nm 处的总透过率随着 Al_2O_3 含量的增加(0→90%)而下降(82.6%→23.6%)。Al_2O_3-YAG:Ce 陶瓷荧光体的激发和发射强度随 Al_2O_3 含量的增加先增大后减小。当 Al_2O_3/Al_2O_3-YAG:Ce 的质量比为 70%时, 陶瓷荧光体在室温下的热导率高达 25.7 $W\cdot m^{-1}\cdot K^{-1}$, 且表现出最高的发射强度。当采用功率密度为 20 $W\cdot mm^{-2}$ 的蓝光二极管泵浦 70% Al_2O_3-YAG:Ce 复相陶瓷荧光体时, 可获得 3724 lm 的高光通量和 239.4 $lm\cdot W^{-1}$ 的高流明效率。此外, 当功率密度从 1 $W\cdot mm^{-2}$ 增大到 20 $W\cdot mm^{-2}$ 时, 流明效率仅下降 10.5%, 光通量持续增加且未出现发光饱和。上述结果显示, Al_2O_3-YAG:Ce 复相陶瓷荧光体具有良好的发光效率和热稳定性, 将在高功率激光照明中具有广阔的应用前景。

关 键 词: Al_2O_3-YAG:Ce; 激光照明; 流明效率; 组分设计; 性能优化

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