RESEARCH ARTICLE

High Luminous Efficacy and Thermal Stability of LuAG:Ce Phosphor Ceramics with Porosity for High-brightness Laser Lighting

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Abstract

The generation of high-brightness white light in laser-illuminated devices is achieved by exciting yellow-green phosphors with a blue laser source. This configuration offers advantages such as high energy density and strong central luminous intensity. Among luminescent materials, LuAG:Ce phosphor ceramics (PCs) are notable for their high thermal stability and luminous output. In this study, LuAG:Ce PCs were produced from co-precipitated monophase nanopowders. The effects of vacuum sintering and air annealing temperatures on their porosity and luminescent properties were investigated. The sample, sintered at 1550°C and subsequently annealed at 1350°C, exhibited 3.5 vol.% porosity, highest room-temperature emission intensity in the series, and retained 96% of that intensity upon heating to 250°C. Under 450 nm laser-diode excitation, the sample achieved a luminous efficacy of 229 lm·W^{-1} while maintaining excellent thermal stability. These results indicate that introducing controlled porosity to act as light-scattering centers in LuAG:Ce ceramics is a viable materials-design strategy for enhancing the luminous performance of laser-driven lighting devices.

Keywords: LuAG:Ce, co-precipitation, ceramics, porosity, luminescence properties.

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1 Introduction

Semiconductor illumination, regarded as fourth-generation lighting technology, represents a significant advancement in photonic engineering for 21st-century applications [1–5]. This system comprises two principal components: light-emitting diodes (LEDs) [6–8] and laser diodes (LDs) [9]. Although LEDs dominate general lighting, their use in high-power applications is limited by a non-linear efficiency drop in luminous output with increasing current density [10–13]. In contrast, LDs provide superior performance for high-power operation, offering stable high-current operation, efficient photon extraction, nanosecond response times, wide operating ranges, and compact form factors. These advantages enable applications in automotive lighting, medical devices, and digital projection [14–16]. Conventional phosphor converters, which disperse phosphors in organic epoxy or silicone resins, are unsuitable for high-brightness laser illumination because they undergo severe thermal degradation under high-power laser excitation [3, 17]. This limitation has driven the development of advanced luminescent materials that combine enhanced thermal dissipation with improved resistance to laser-induced damage [18, 19].

Among advanced luminescent materials, including inorganic phosphors [13, 20], phosphor crystals [21, 22], phosphor ceramics (PCs) [23–27], and phosphor in glasses (PiGs) [28–33], PCs are notable for their superior thermal conductivity and high thermal stability. PCs have attracted considerable research attention due to their high luminous output, established fabrication routes, cost-effectiveness, and ability to integrate multiple phosphor components. Among these, LuAG:Ce and YAG:Ce ceramics [25] exhibit excellent thermal stability and are among the primary materials currently under investigation [26]. PCs are typically used in two application modes—transmissive and reflective—and are produced in either transparent or opaque forms to fit these configurations [34]. Because transparent PCs often have low excitation-light utilization efficiency [40], researchers have introduced optical scattering centers, such as secondary phases and pores, to enhance excitation-light harvesting in these materials [35, 36]. Incorporating an appropriate secondary phase can improve thermal conductivity. However, it also increases internal scattering (predominantly Rayleigh scattering), which may reduce central luminous intensity. In porous ceramics, Mie scattering associated with the pores can instead enhance central intensity [37]. Computational studies indicate that controlled porosity creates refractive-index discontinuities that act as effective photon scattering centers [36]. Raukas et al. [8] were among the first to propose exploiting residual porosity as optical scattering centers and analyzed its effect on photon-extraction efficiency under low-power-density blue LED excitation. Zheng et al. [16] produced YAG:Ce ceramics with approximately 15% porosity, achieving improved luminous homogeneity and high radiance through optimized photon confinement. Zhang et al. [38] fabricated LuAG:Ce ceramics with variable porosity by adjusting sintering temperatures and reported that samples sintered at 1650°C with 2.9% porosity exhibited the highest emission intensity, reaching a luminous efficacy (LE) of 200 $lm{\cdot}W^{-1}$ under 450 nm LD excitation. Yang et al. [39] similarly observed that increased internal porosity in LuAG:Ce ceramics enhanced central light intensity in reflective, laser-transparent modules. Nevertheless, the relationships among pore size, shape, distribution, and fluorescent performance under high-flux laser irradiation remain insufficiently understood and require systematic investigation.

This study presents a sintering-temperature-based porosity engineering approach for LuAG:Ce PCs and demonstrates that porosity, controlled within the experimentally realized sintering temperature range, can serve as an effective variable to enhance luminous performance in laser-driven lighting devices.

2 Experimental

The LuAG:Ce nanopowders were synthesized using the co-precipitation method. $Lu(NO_3)_3$ and $Ce(NO_3)_3$ solutions were synthesized by dissolving Lu_2O_3 (99.99%, Shanghai Jingyun Material Technology Co., Ltd., China) and CeO_2 (99.999%, Changting Golden Dragon Rare-Earth Co., Ltd., China) in hot high-purity nitric acid, respectively. The $Al(NO_3)_3$ solution was prepared by dissolving $Al(NO_3)_3 \cdot 9H_2O$ (99.0%, Sinopharm Chemical Reagent Co., Ltd., China) in deionized water. The stoichiometric mole ratio of metal nitrates was mixed according to the chemical formula LuAG:0.55 at%Ce and diluted to a 0.5 M solution. The precipitant solution was obtained by dissolving the ammonium hydrogen carbonate (AHC) (analytical grade, Aladdin Scientific Co., China) in deionized water and diluted to a 1.5 M solution. Ammonium sulfate (99.0%, Sinopharm Chemical Reagent Co., Ltd., China) was added to the precipitant solution and

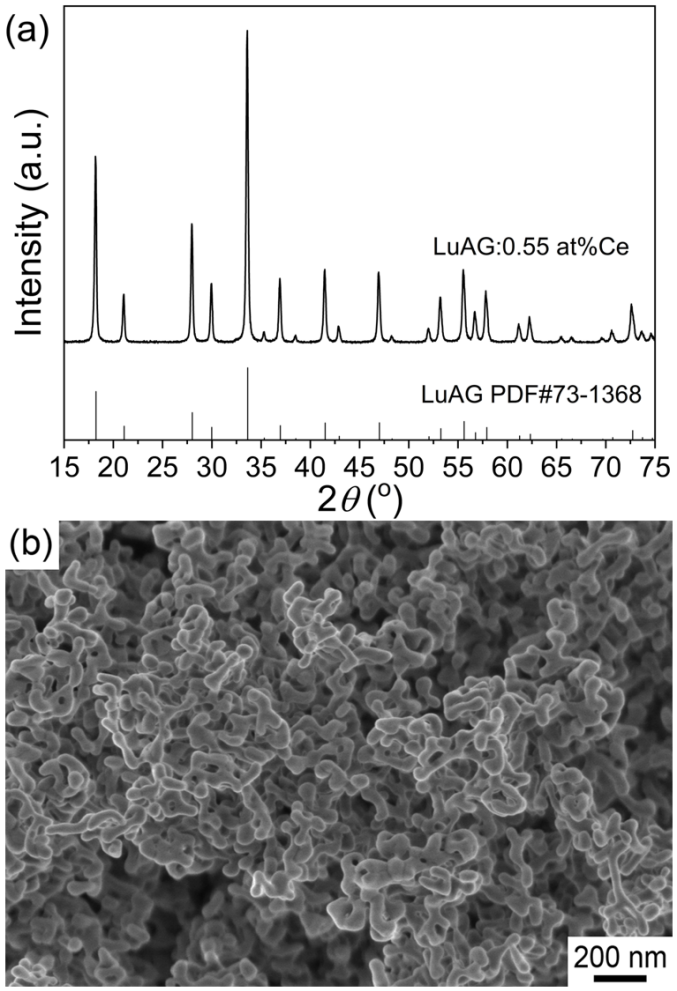


Figure 1. (a) XRD pattern and (b) FESEM micrograph of LuAG:Ce nanopowders calcined at 1100°C for 4 h in air.

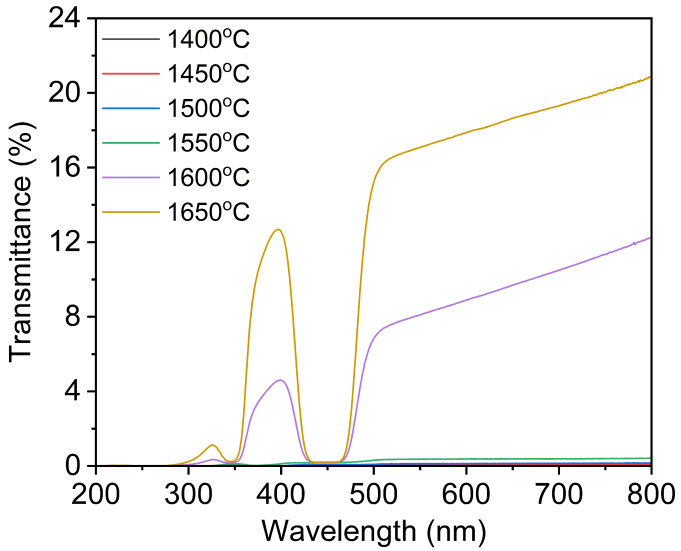


Figure 2. In-line transmittance of LuAG:Ce ceramics (0.4 mm thick) annealed at 1100°C for 5 h in air.

acted as a dispersant. Reverse-strike titration was performed by dripping the mixed metal nitrates into the AHC solution at a rate of 20 mL/min at room temperature (RT). After aging for 1 h, the resulting suspension was washed three times with deionized water and rinsed twice with absolute ethanol by repeated dispersion and centrifugal filtration. Then, the precursor was dried at 70°C for 48 h, sieved through a 200-mesh screen, and calcined at 1100°C for 4 h. The dry-pressed green bodies using the synthesized nanopowders were vacuum-sintered at different temperatures (1400-1650°C) for 3 h, followed by air annealing at 1350 or 1450°C for 10 h. For further studies, the ceramic samples were ground and mirror-polished on both sides to thickness of 0.4 mm.

3 Results and Discussion

The phase identification of the samples was performed by X-ray diffraction (XRD; Ultima IV, Rigaku, Japan) in the 2θ range of 15-75° using nickel-filtered Cu Kα radiation. Micrographs of the thermally etched surfaces of ceramics (1350°C×3 h in air) were characterized by field emission scanning electron microscopy (FESEM; SU8220, Hitachi, Japan). The relative density of the LuAG:Ce PCs is calculated according to the Archimedes principle and the porosity is derived from the relative density. The theoretical densities of these PCs are obtained according to the XRD pattern of LuAG:Ce nanopowders calcined at 1100°C for 4 h in air. The homemade multi-function combined fluorescence spectrum test system (SicOmni-I) was used for the UV-VIS fluorescence test. The system was equipped with a VX-XBO 150 W xenon lamp, an Omni-λ3007 (Zolix, China) monochromator, and a PMTH-S1-CR131 photomultiplier tube. The spectral wavelength was corrected by an LHM254 mercury lamp. Variable temperature photoluminescence (PL) spectroscopy was measured using a FluoroMax-4 spectrofluorometer (HORIBA, Japan). The related RT LE, luminous flux (LF), and electroluminescent properties were measured by an integrating sphere connected to a CCD spectrometer OHSP-350 (HOPOCOLOR, China).

The XRD pattern and FESEM micrograph of LuAG:0.55 at%Ce nanopowders after air calcination are presented in Figure 1. All diffraction peaks match those of the $Lu_3Al_5O_{12}$-phase standard reference (PDF#73-1368), with no detectable secondary phases. The sharp peaks and high diffraction peak intensities in the XRD pattern indicate their high crystallinity. The powders exhibit a distinct elongated morphology with an average particle size of approximately 70 nm. XRD and FESEM investigations indicate that parameters of

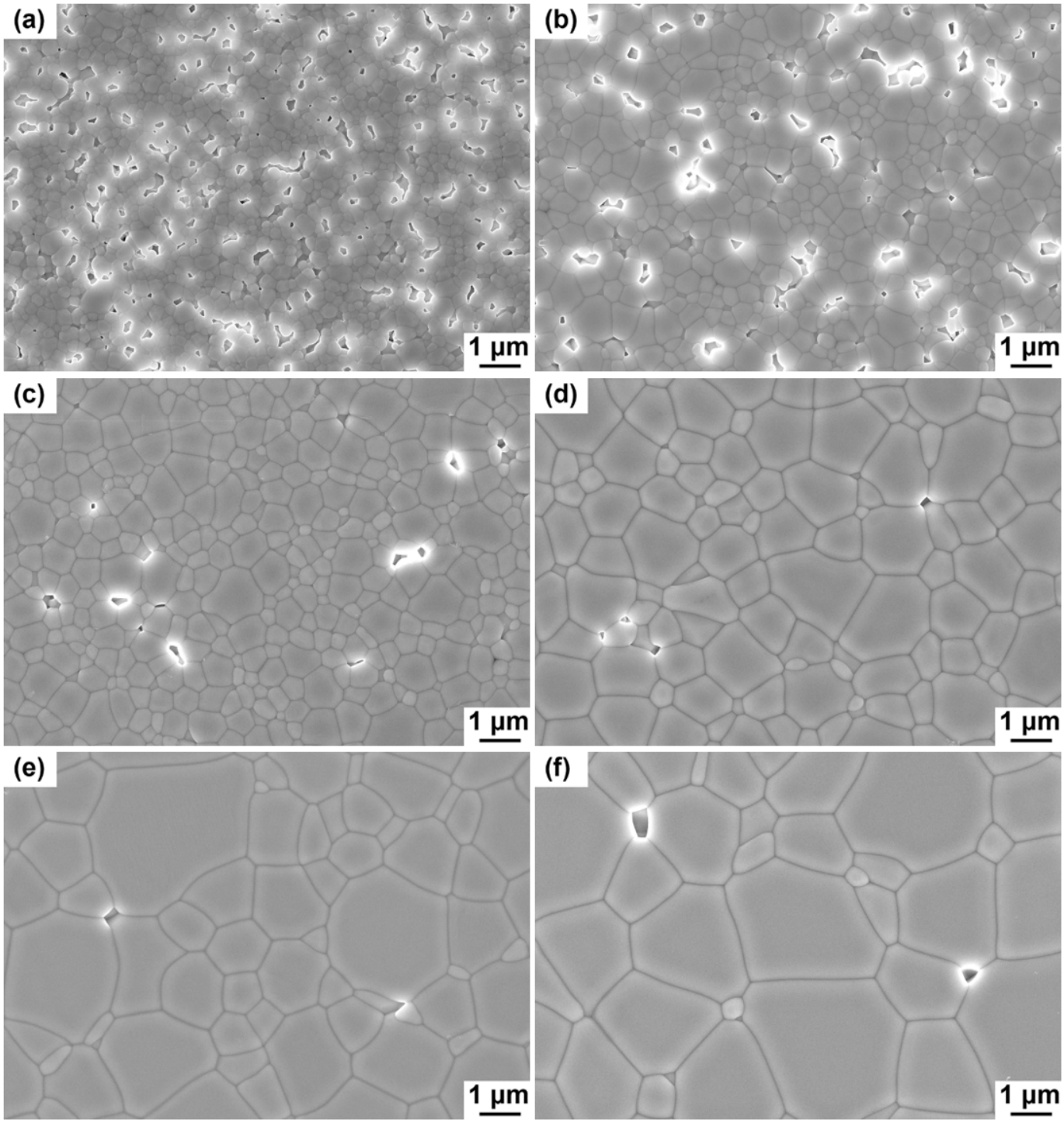


Figure 3. FESEM micrographs of LuAG:Ce ceramics vacuum-sintered at different temperatures for 3h: (a) 1400°C, (b) 1450°C, (c) 1500°C, (d) 1550°C, (e) 1600°C, (f) 1650°C.

nanopowders synthesized by co-precipitation provide favorable preconditions for subsequent optical ceramic preparation during sintering.

In-line transmittance of LuAG:Ce ceramics are shown in the Figure 2. It is observed that when the vacuum sintering temperature of ceramics reaches 1400–1550°C, the ceramic's in-line transmittance approaches zero. At a sintering temperature of 1650°C, the ceramic achieves an in-line transmittance of 20.9% at 800 nm. Notably, two absorption bands observed at 350 nm and between 400–500 nm correspond to electron transitions $4f \rightarrow 5d_1$ and $4f \rightarrow 5d_2$ from the ground state to the excited state energy levels of Ce^{3+}, respectively.

The SEM micrographs of thermally etched surfaces of LuAG:Ce ceramics are shown in Figure 3(a-f).

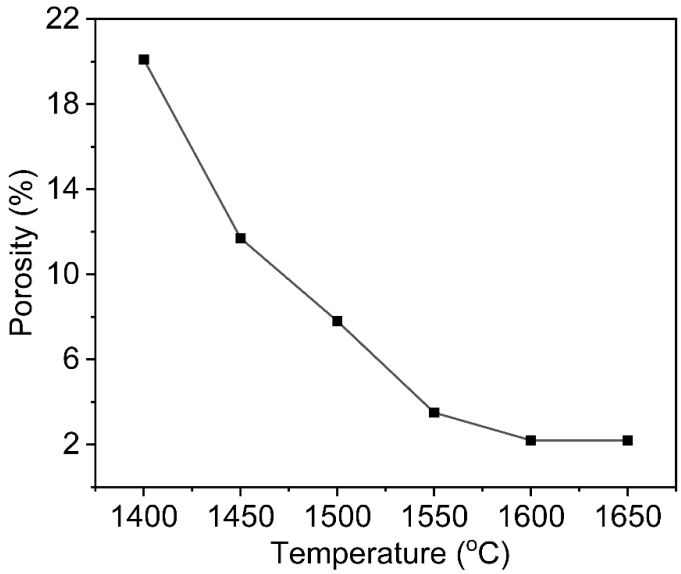


Figure 4. Porosity of LuAG:Ce ceramics vacuum-sintered at different temperatures for 3h.

The average grain size of the samples increases significantly with sintering temperature from 431 nm to 662 nm, 1440 nm, 1890 nm, 1900 nm, and 2530 nm. The number of micropores decreases with increasing temperature, which implies that the porosity of the ceramics gradually decreases (Figures 3 and 4). However, when the temperature increases from 1600 to 1650°C, the average grain size increases, while the porosity remains almost the same (2 vol.%). The increased sintering temperature accelerates the grain boundary migration, leading to an increase in grain size. However, the rapid movement of grain boundaries engulfs pores and forms closed pores within grains.

The SEM micrographs of the fracture surfaces of LuAG:Ce ceramics are shown in Figure 5(a-f). The micrographs reveal that the shape and size of pores on the fracture surfaces correspond to those on the ceramic surfaces. The fracture surface micrographs exhibit the same variation patterns in grain size and porosity as those observed on the ceramic surfaces.

The effects of porosity on LE should be considered from both aspects of incident and converted light. It has been found that scattering centers can enhance both light absorption by activators and light extraction from ceramics in applications of PCs [38].

In LuAG:Ce PCs, low-porosity samples (sintered at ≥1600°C) exhibit limited light absorption by luminescent centers, as incident blue light transmits directly through the bulk phosphor with minimal interaction (the lower panel in Figure 6). In contrast, higher-porosity ceramics demonstrate distinct behavior (the down panel in Figure 6): controlled scattering amplifies photon-Ce^{3+} activator interactions, significantly enhancing blue light absorption. Furthermore, porosity-induced scattering improves light extraction efficiency by redirecting photons toward emission surfaces. However, when the porosity of the ceramics is high enough, light has little chance to pass through the ceramics.

The present study aims to assess the suitability of LuAG:Ce ceramics for high-power laser illumination systems. To this end, the luminous performance of the samples obtaining at different post-annealing temperatures was measured under 450-nm laser excitation. A reflection mode was applied. As demonstrated in Figure 7, under 1 $W{\cdot}mm^{-2}$ power laser excitation, the LE of ceramics at varying annealing temperatures exhibited an initial increase followed by a subsequent decrease with increasing sintering temperature. However, it is evident that the LE of samples annealed at 1350°C is higher than those annealed at 1450°C. This phenomenon can be attributed to the fact that an increase in annealing temperature promotes the oxidation of Ce^{3+} to Ce^{4+} within the ceramics. Consequently, the number of luminescent centers is reduced, thereby decreasing the LE. LuAG:Ce PCs vacuum-sintered at 1550°C and subsequently air annealed at 1350°C exhibited a maximum LE value of 229 $lm{\cdot}W^{-1}$.

The absorption spectra of LuAG:Ce ceramics sintered at 1550°C and post-annealed at different temperatures were measured. As shown in Figure 8, the increase of annealing temperature is accompanied by an increase in the charge-transfer (CT) absorption band associated with Ce^{4+}. This observation suggests a reduction in the concentration of Ce^{3+} luminescent centers (due to partial oxidation to Ce^{4+}), which leads to a decrease in the ceramics' overall LE (Figure 7).

Figure 9(a) demonstrates the PL spectral characteristics of LuAG:Ce PCs vacuum-sintered at different temperatures for 3 h and annealed at 1350°C for 10 h. A broad emission band from 470–650 nm is observed, originating from the radiative transition of Ce^{3+} from 5d to the 4f ground state. When the sintering temperature increases from 1400°C to 1650°C, the PL intensity first increases and then decreases from 1600°C. This phenomenon suggests that appropriate porosity can act as the scattering centers to increase blue light absorption and yellow-green light extraction from the ceramic samples, allowing the PCs to achieve

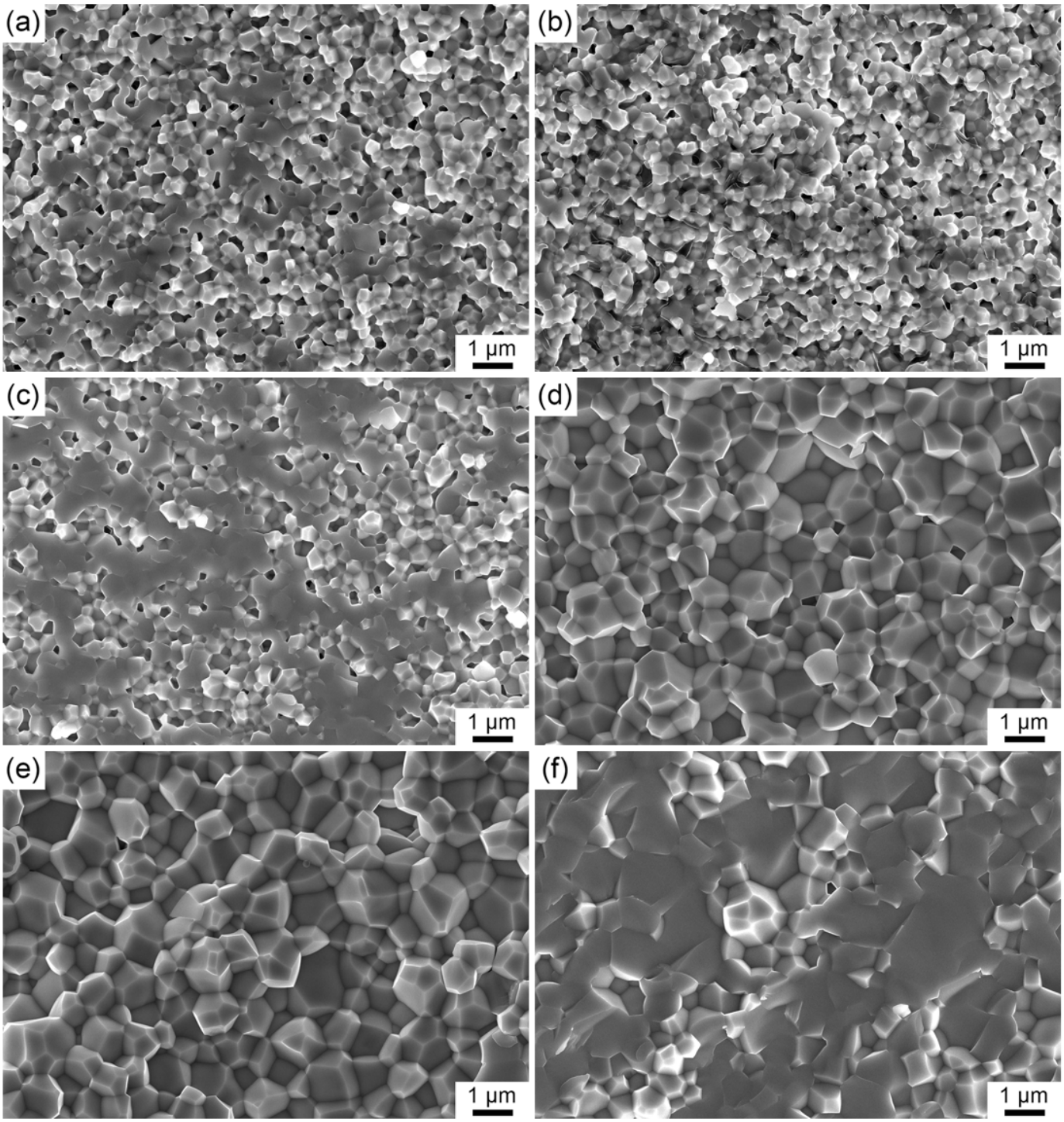


Figure 5. FESEM micrographs of the fracture surfaces of LuAG:Ce ceramics vacuum-sintered at different temperatures for 3h: (a) 1400°C, (b) 1450°C, (c) 1500°C, (d) 1550°C, (e) 1600°C, (f) 1650°C.

good LE. The PL intensity of the sample with a 3.5 vol.% porosity (sintered at 1550°C) is the highest (Figure 9).

Figure 10 demonstrates the temperature-dependent PL spectral properties of the ceramic sample sintered at 1550°C for 3h and annealed at 1350°C for 10h. The decrease in PL intensity is only 4% when the temperature increases to 250°C, suggesting that the prepared ceramic samples possess desirable thermal stability. In general, PCs have higher thermal conductivity than phosphor powders and therefore transfer heat faster, which ultimately reduces the rate of phonon-assisted nonradiative transitions [5, 41].

The LE values of the LuAG:Ce ceramic sample sintered at 1550°C and annealed at 1350°C decreases with the increase in incident blue LD power density,

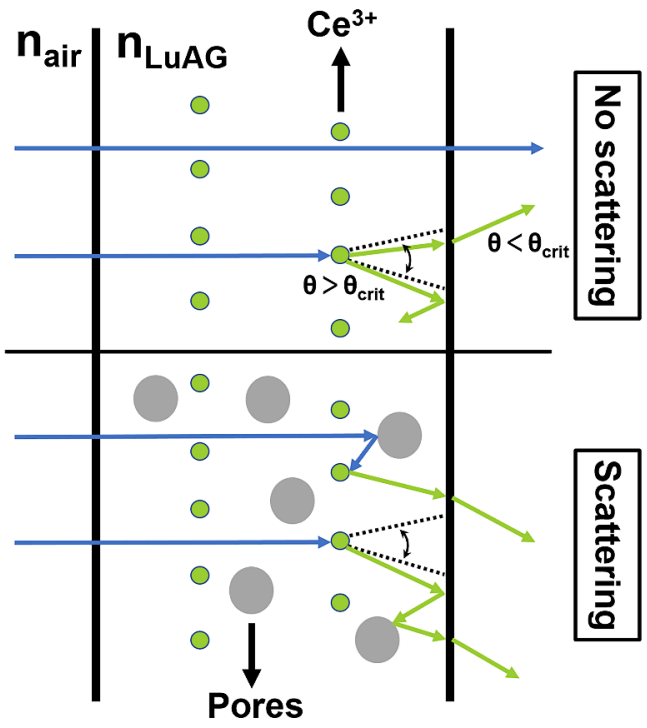


Figure 6. Schematic diagram of light propagation in LuAG:Ce PCs with and without scattering centers.

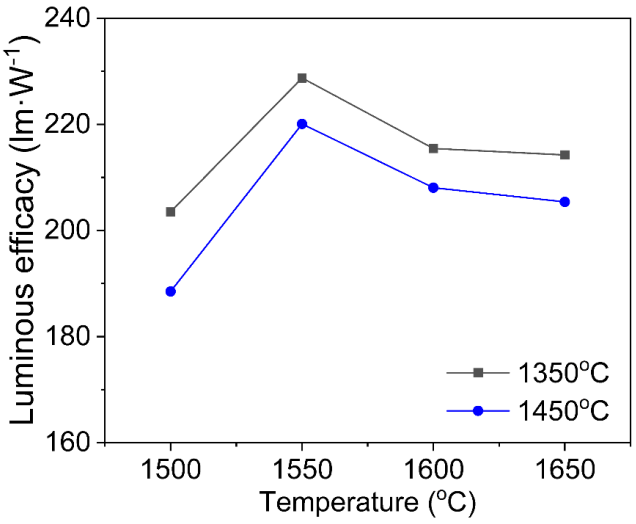


Figure 7. LE values of LuAG:Ce ceramics vacuum-sintered at different temperatures and air annealed at 1350 or 1450°C for 10h (under excitation of 1 W·mm^{-2} blue LD).

while the LF increases gradually and saturates at 18 W·mm^{-2} (Figure 11(a)). The emission intensity of the ceramic samples increases with the increase of power excitation, while the luminous intensity does not decrease substantially under the higher incident LD power of 20 W·mm^{-2} (Figure 11(b)). These dependences are consistent with those shown in Figure 10 and indicate that the sample has a good thermal performance.

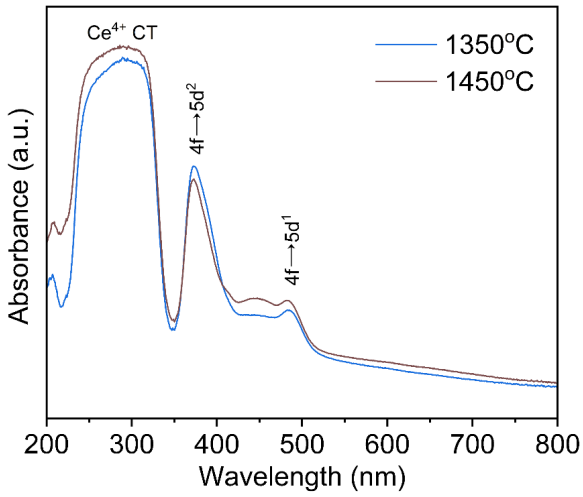


Figure 8. Absorption spectra of LuAG:Ce PCs vacuum-sintered at 1550°C for 3h and annealed at different temperatures for 10h.

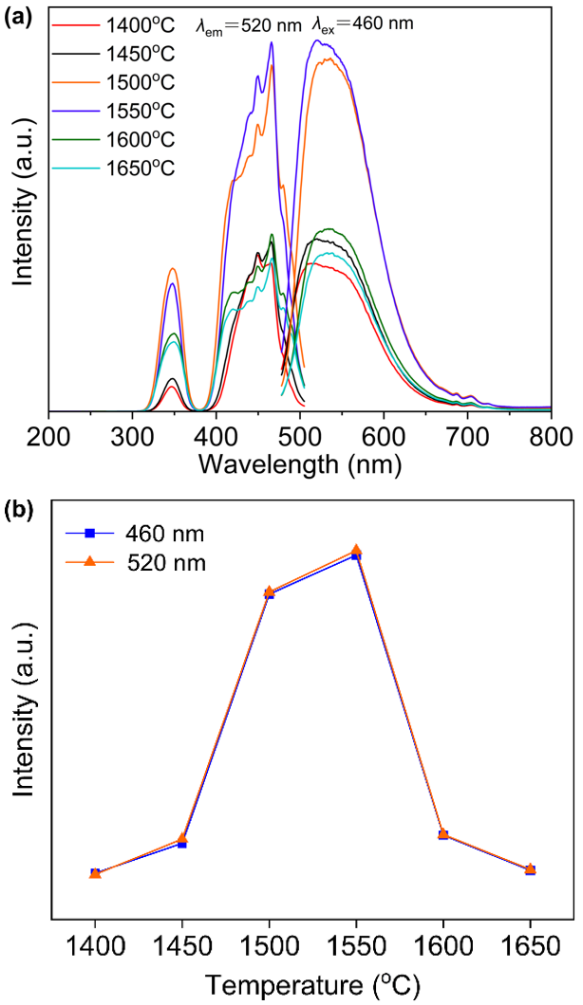


Figure 9. (a) PL/PLE spectra and (b) PL peak intensities of LuAG:Ce PCs vacuum-sintered at different temperatures for 3h and annealed at 1350°C for 10h.

4 Conclusions

It was shown, the LuAG:Ce nanopowders prepared via the co-precipitation method exhibits excellent

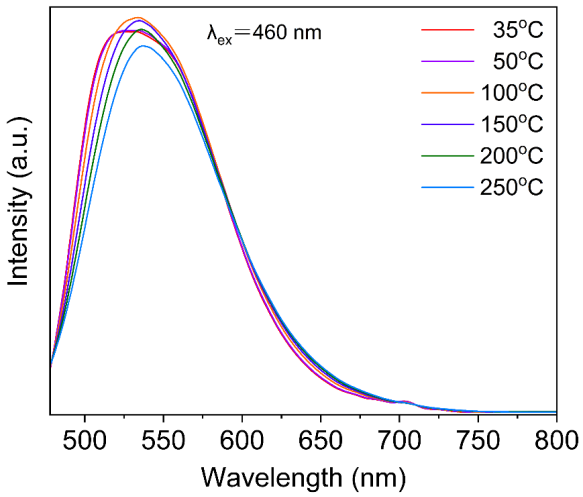


Figure 10. Temperature-dependent PL spectra of LuAG:Ce PCs sintered at 1550°C and annealed at 1350°C.

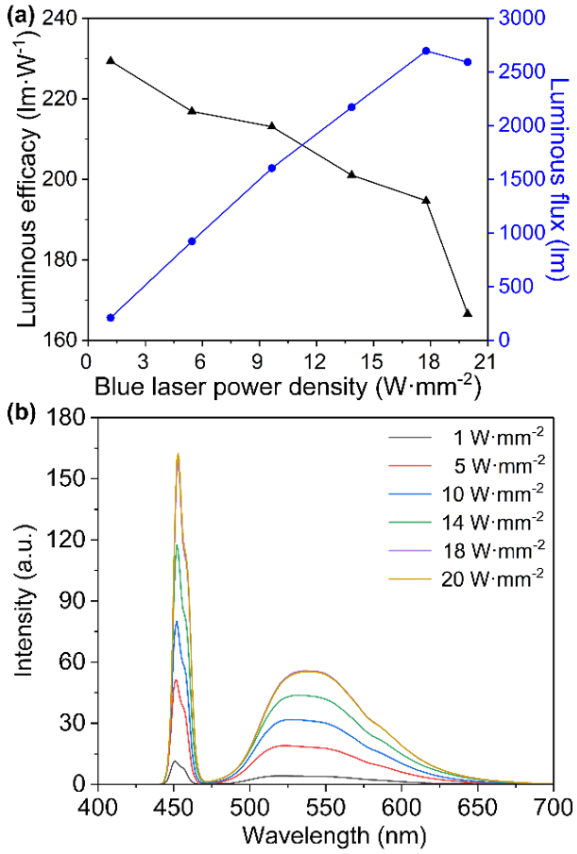


Figure 11. (a) Variation of luminous efficacy and luminous flux with blue laser power density and (b) Emission spectra of LuAG:Ce ceramics (0.4 mm thick) sintered at 1550°C for 3 h in vacuum and then annealed at 1350°C for 10 h in air, under blue light excitation with different incident powers.

sintering activity, enabling the fabrication of PCs with varying porosities at relatively low vacuum sintering temperatures. The influence of vacuum sintering and air annealing temperatures on the microstructure and luminescent properties of LuAG:Ce PCs were systematically studied. LuAG:Ce ceramics with a porosity of 3.5% vacuum sintered at 1550°C for 3 h and air annealed at 1350°C for 10 h exhibit the highest room-temperature luminous intensity among all samples, and LuAG:Ce ceramics show only 4% degradation when the temperature increases from 35°C to 250°C. Under excitation of 1 W·mm^{-2} blue LD, this sample exhibited a remarkable LE of 229 lm·W^{-1} while maintaining excellent thermal stability. An increasing trend of LF without showing luminescence saturation is observed under density up to ~18 W·mm^{-2}. This research indicates that the strategic incorporation of porosity within single phase ceramics can remarkably enhance the luminous performance of laser lighting devices, as also establishes the foundation for subsequent investigations in the context of the development of multiphase PCs.

Data Availability Statement

Data will be made available on request.

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Conflicts of Interest

The authors declare no conflicts of interest.

AI Use Statement

The authors declare that no generative AI was used in the preparation of this manuscript.

Ethical Approval and Consent to Participate

Not applicable.

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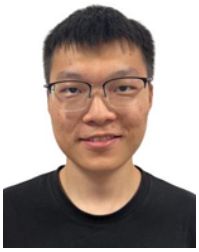
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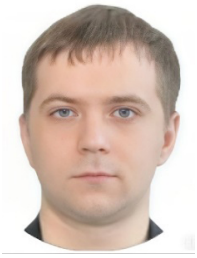
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