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Letter

Effect of La_2O_3 on microstructures and laser properties of Nd:YAG ceramics

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ABSTRACT

Transparent 1.0 at.% Nd:YAG ceramics were fabricated by vacuum sintering technology using commercial α-Al_2O_3, Y_2O_3 and Nd_2O_3 powders as raw materials. Influence of La_2O_3 additions (0–1.2 wt%) on microstructures and optical properties of Nd:YAG ceramics was investigated. The results indicate that the optical properties of Nd:YAG ceramics with 0.4 wt% La_2O_3 are superior than those of undoped-Nd:YAG ceramics. When the amount of La_2O_3 is 0.8 wt%, the specimen has the highest transmittance in the region from 400 nm to 1100 nm. No pores or other defects are found in or between the grains. However, residual inclusions along grain boundaries and pores are easily generated by adding excessive La_2O_3 (1.2 wt%). The Nd:YAG ceramics are pumped by a diode laser to study the laser properties. The slope efficiency and threshold of Nd:YAG ceramics with optimum addition of La_2O_3 (0.8 wt%) are 41.1% and 2.9 W, respectively, which are the best laser oscillation results among all the specimens.

1. Introduction

As a solid-state laser material, neodymium-doped yttrium aluminum garnet (Nd:YAG) single crystals fabricated by the Czochralski method has been widely used as solid in laser systems for industrial, medical, scientific, and military purposes [1,2]. However, the growing technique for Nd:YAG single crystals requires great skills, the requirement of expensive Ir crucibles and the difficulty in growing large size crystal with high Nd^{3+} doping concentration has prevented their further applications [3]. In 1995, Ikesue et al. fabricated transparent Nd:YAG ceramics with the required optical properties for laser applications [4,5]. Since then, Nd:YAG transparent ceramics have attracted much attention because the optical properties have been improved greatly and the highly efficient laser oscillations have been obtained.

Compared with single crystals, Nd:YAG ceramics have several advantages, such as lower cost, large size, high concentrations doping, multifunctional composite structure, and so on [6,7]. Thus, tremendous efforts have been made to develop synthesis methods for YAG ceramics [8–15]. In order to fabricate high-transparency Nd:YAG ceramics, pores should be eliminated as much as possible [16]. If the grain size is too large, it is difficult to remove the residual pores because there are less grain boundaries for pores release. So it is better to find a way to control the grain growth rate. Some reports [17,18] show that the abnormal grain growth can be inhibited by adding sintering aids such as MgO and SiO_2. Unfortunately, a few residual pores are still observed, the optical qualities of Nd:YAG ceramics are a little inferior to those of single crystals due to the residual pores in the ceramics act as optical scattering centers [5]. Lanthana (La_2O_3) is a sintering aid which has been successfully applied to fabricate Y_2O_3 ceramics [19,20]. The addition of La_2O_3 is helpful to promote desification and decrease sintering temperature, and then decrease the grain boundary diffusion coefficient. The La_2O_3-doped Y_2O_3 ceramics display pore-free microstructures and high optical qualities [21]. Therefore, it is possible to improve the optical properties of Nd:YAG ceramics by doping La_2O_3. In this work, Nd:YAG ceramics were fabricated by solid-state reaction and vacuum sintering method using La_2O_3 as a sintering aid. The effect of various amount of La_2O_3 on the microstructures and optical properties of Nd:YAG ceramics was investigated.

2. Experimental procedures

2.1. Fabrication of Nd:YAG ceramics

High-purity powders of α-Al_2O_3 (≥99.99%, Alfa Aesar Company, USA), Y_2O_3 (≥99.99%, Alfa Aesar Company, USA), Nd_2O_3 (≥99.99%, Alfa Aesar Company, USA) and La_2O_3 (≥99.99%, Alfa Aesar Company, USA) were used as starting materials. These powders were mixed according to the stoichiometric $Y_{2.97}Nd_{0.03}Al_5O_{12}$ with different contents of La_2O_3 and ball-milled with the high-purity Al_2O_3 balls for 10 h in ethanol using tetraethylorthosilicate (TEOS) as sintering aid. The mass fractions of La_2O_3 in the resultant Nd:YAG ceramics were controlled to 0 wt%, 0.4 wt%, 0.8 wt% and 1.2 wt%, respectively. The mixture of the powders was dried and sieved through a 200-mesh screen. After removing the organic component by calcining at 800 °C for 4 h, the powders were dry-pressed under 20 MPa into Ø 30 mm × 7 mm disks and finally cold-isostatically pressed under 250 MPa. Green compacts were sintered at 1730 °C for 20 h under vacuum (1.0×10^{-3} Pa), after that, the pellets were annealed

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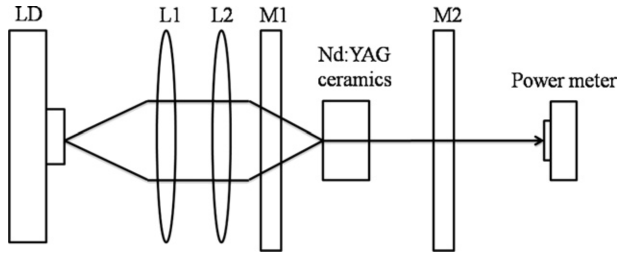

Fig. 1. Schematic diagram of the laser experimental setup.

at 1450 °C for 20 h in air and mirror polished on both surfaces. The Nd:YAG ceramics were obtained, and named as specimens A, B, C and D, respectively.

The microstructures of specimens were observed by electron probe micro-analyzer (EPMA, Model JXA-8100, JEOL, Japan). The transmittance spectra were measured with a spectrophotometer (Model U-2800, Hitachi, Japan). The photo-luminescence spectra were measured at room temperature by spectrofluorometer (Model Fluorolog-3, Jobin Yvon, France), with an 808 nm diode laser used as pump source.

2.2. Laser experiment

The experimental setup for oscillation is shown in Fig. 1. The Nd:YAG ceramics are processed into shape of 3 mm × 3 mm × 6 mm and mirror polished on both surfaces and coated on two parallel end facets. The input facets are high transmittance coated at 808 and 1064 nm, while the output facets are antireflection coated at 808 nm. A 15 W ring-shaped laser diode (LD) with the emission wavelength of 808 nm is used as pump source. The laser cavity consists of two flat mirrors (M1 and M2), where M1 is antireflection-coated at 808 nm and high-reflection at 1064 nm as the input coupler (IC), and M2 is output coupler (OC) with transmittance of 5% at 1064 nm.

3. Results and discussion

Fig. 2 shows a photograph of mirror polished Nd:YAG ceramics sintered at 1730 °C for 20 h after annealing at 1450 °C for 10 h. All specimens exhibit good transparency. Words behind it can be read clearly. As can be seen from Fig. 3, the transmittances of specimens A, B, C and D are all above 82% at the lasing wavelength of 1064 nm. According to the Rayleigh's equation, the scattering intensity increases with decreasing of wavelength [22]. The optical transmittance is 79.7% for specimen A, 81.4% for specimen B, 82.5% for specimen C and 79.6% for specimen D at 400 nm, respectively. The transmittance of specimen C is the best of all in the region from

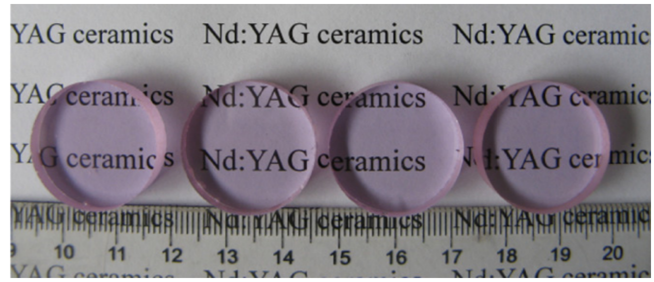

Fig. 2. Photograph of specimens (5 mm thickness) with various contents of La_2O_3, (a) without La_2O_3, (b) 0.4 wt%, (c) 0.8 wt%, and (d) 1.2 wt%.

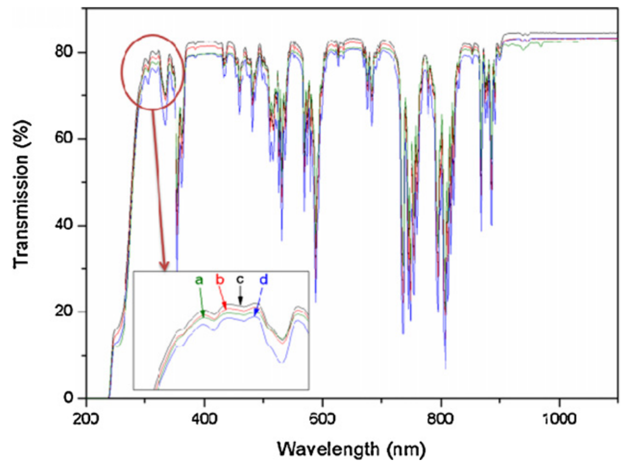

Fig. 3. Optical transmissions of specimens with various contents of La_2O_3, (a) without La_2O_3, (b) 0.4 wt%, (c) 0.8 wt%, and (d) 1.2 wt%.

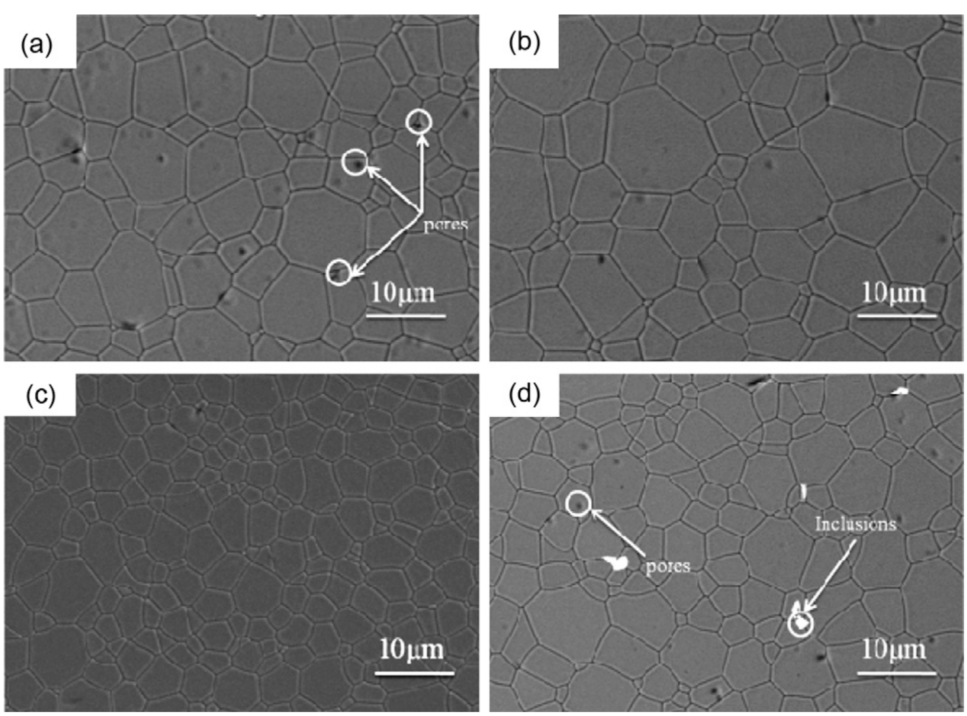

Fig. 4. SEM of specimens with various contents of La_2O_3, (a) without La_2O_3, (b) 0.4 wt%, (c) 0.8 wt%, and (d) 1.2 wt%.

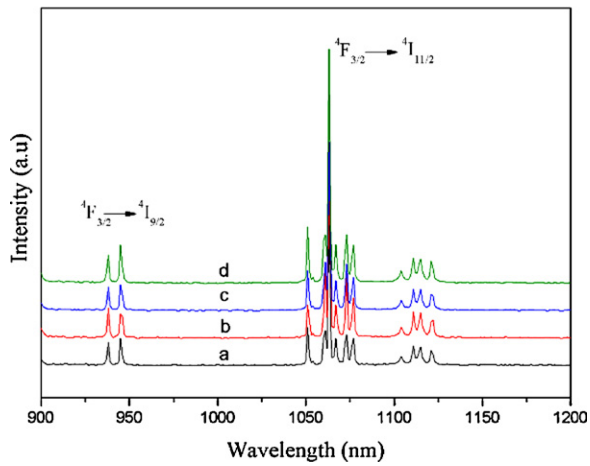

Fig. 5. Photoluminescence spectra of specimens with various contents of La_2O_3, (a) without La_2O_3, (b) 0.4 wt%, (c) 0.8 wt%, and (d) 1.2 wt%.

400 nm to 1100 nm, which is very close to that of single crystal [4]. The result indicates that the optimum addition of (0.8 wt%) La_2O_3 will help to improve the optical properties of Nd:YAG ceramics.

In order to detect the main reason for decreasing of the transmittance of specimens at 400 nm, the micrographs of specimens with different quantities of La_2O_3 are observed by EPMA (Fig. 4). If there is no La_2O_3 as a sintering aid, a few pores are observed both at the grain boundaries and inner grains (Fig. 4(a)). For La_2O_3 dopant of 0.4 wt%, the amount of pores decreases in the Nd:YAG ceramics (Fig. 4(b)), and for 0.8 wt%, no pores or other defects can be found (Fig. 4(c)). The average grain size of specimen C with pore-free microstructures is smaller than that of specimen B. It is because that La_2O_3 is prone to segregate at the grain boundaries, which will decrease the grain growth kinetics. As the grain growth rate is slower than the pore diffusion rate, pores can be removed easily from the specimen C. However, as can be seen from Fig. 4(d), some residual inclusions and apparent pores are formed at grain boundaries because of the addition of excessive La_2O_3 (1.2 wt%). This is detrimental to the optical properties of Nd:YAG ceramics (Fig. 3). Consequently, the addition of La_2O_3 should be accurately controlled.

The photoluminescence spectra of Nd:YAG ceramics with various La_2O_3 contents are shown in Fig. 5. From 900 nm to 1200 nm, there are two emission bands centered at 946 nm and 1064 nm, which are corresponding to the transition from $^4F_{3/2} \rightarrow {}^4I_{9/2}$ and $^4F_{3/2} \rightarrow {}^4I_{11/2}$ of Nd^{3+} ions, respectively. No obvious wavelength redshift has been observed with increasing concentrations of the La_2O_3. The fluorescent line widths for Nd:YAG ceramics with 0 wt%, 0.4 wt%, 0.8 wt% and 1.2 wt% La_2O_3 are almost identical. It means the addition of small amount La_2O_3 has no effect on the spectral properties of Nd:YAG ceramics despite of the slight lattice distortion caused by its doping.

Fig. 6 shows laser output versus pump power for Nd:YAG ceramics at 1064 nm. With 13.6 W of maximum absorbed pump power, laser output power of specimens B and C are 3.3 W and 4.4 W respectively. The threshold for both specimens B and C is 2.9 W. The slope efficiency of specimen B is 30.8%, which is 10.3% lower than that of specimen C (41.1%). The laser output of specimens A and D cannot be achieved, the main reason is that there are some residual pores or other defects in the two specimens (Fig .4(a and d)). Compared with the previous reports, it is also found that the laser properties of specimen C are inferior to those of single crystals [5]. These may attribute to losses mainly caused by grain boundaries and some residual defects (cannot be detected) in the Nd:YAG

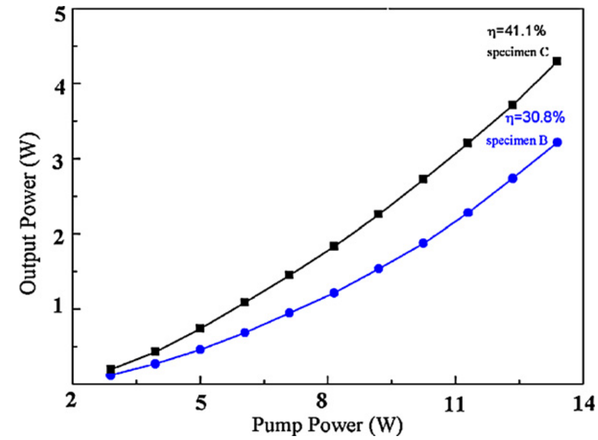

Fig. 6. Laser output at 1064 nm versus pump power.

ceramics. The detailed relation between microstructures and scattering loss will be further studied.

4. Conclusions

Transparent Nd:YAG ceramics were fabricated by solid-state reaction and vacuum sintering using La_2O_3 as sintering aid. The small amount of La_2O_3 helps to control abnormal grain growth, and refine the microstructures of Nd:YAG ceramics. The maximum refinement in microstructures is obtained with the optimum addition (0.8 wt%) of La_2O_3. The specimen C with average grain size of about 8 μm has a fine microstructure. No pores or other defects appear in or between the grains. The optical properties of specimen C are superior to those of the other three specimens in the region from 400 nm to 1100 nm. The slope efficiency and threshold for specimen C are 41.1% and 2.9 W, respectively, which are the best laser oscillation results in all the specimens.

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