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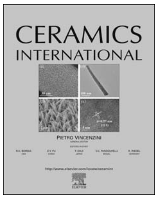

Post-treatment of nanopowders-derived Nd:YAG transparent ceramics by hot isostatic pressing

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ABSTRACT

Neodymium doped yttrium aluminum garnet (Nd:YAG) transparent ceramics were fabricated from Nd:YAG nanopowders synthesized via a reverse precipitation method by vacuum sintering and successive hot isostatic pressing (HIP) post-treatment. The powders obtained by calcining the precursor at 1100 °C for 4 h and then ball milling for 2 h with 0.5 wt% TEOS as sintering aid were used to fabricate Nd:YAG ceramics. The green bodies were vacuum sintered at 1500–1800 °C for 10 h, followed by the HIP at 1600 °C for 3 h in 200 MPa Ar atmosphere. Influence of the calcination temperature on the phase, morphology and particle size evolution of the nanopowders, as well as the optical transparency and microstructure of the obtained Nd:YAG ceramics before and after the HIP post-treatment was investigated in detail. It was found that for the post-treated 1800 °C-vacuum-sintered Nd:YAG ceramic sample, the in-line transmittance increased from 48.0% up to 81.2% at the lasing wavelength of 1064 nm.

1. Introduction

Rare-earth ions doped yttrium aluminum garnet ($Y_3Al_5O_{12}$, YAG) single crystals have been widely used as the host material for solid-state lasers due to the high transmittance, high strength, low creep rates at high temperatures (>1000 °C), high chemical corrosion resistibility, and high mechanical properties [1–4]. However, the fabrication of rare earth doped YAG single crystal, such as Nd:YAG, with laser quality is expensive and has some limitations, for instance it is extremely difficult to produce large-sized Nd:YAG single crystals. Furthermore, doping concentration in the Nd:YAG single crystals is limited due to the small segregation coefficient of the doped Nd^{3+} ions [5]. In 1984, the translucent YAG ceramics were first produced by With and Dijk using a solid-state reaction method and a modified sulfate process [6]. After that in 1995 Ikesue et al. demonstrated that with the help of the solid-state reaction method it is possible to obtain Nd:YAG ceramics with such high quality that it can be used for laser oscillation [7]. Nowadays YAG ceramics have become attractive as a result of their beneficial aspects, such as a relatively short cycle and low cost in manufacture, as well as little limitations in size and shape [8,9]. But it was reported [10,11] that the optical quality of the conventional transparent YAG ceramics is inferior to that of the single crystals, due to the residual pores and second phases in the ceramics acting as light scattering centers. Thus, it is extremely important to eliminate the scattering centers in order to fabricate highly transparent YAG ceramics, which meet exacting requirements of optical applications.

To obtain Nd:YAG ceramics with high optical quality, two methods are generally employed. The frequently-used one is the solid-state reaction sintering (SSRS) method using Nd_2O_3, Y_2O_3, Al_2O_3 powders as starting raw materials [12–15]. The SSRS is a relatively simple way to fabricate Nd:YAG transparent ceramics, however it requires a high temperature and long heating time to achieve high quality of the material [16–18]. It was found that using Nd:YAG nanopowders synthesized by wet chemical methods can overcome the problem [19]. The precipitation method, including homogeneous precipitation, normal co-precipitation and reverse co-precipitation, is the most attractive way to prepare high-quality YAG nano-powders and transparent ceramics [20–23]. Highly transparent Nd:YAG ceramics with the in-line transmission of $>82\%$ were produced by Stevenson et al. from powders synthesized by reverse strike co-precipitation with SiO_2

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as the sintering aid [24]. Ma et al. demonstrated the fabrication of Nd:YAG transparent ceramics with the transmittance higher than 81.5% at 1064 nm by dry pressing, vacuum sintering, and air annealing, using the raw powders synthesized via a modified co-precipitation method with the ethanol–water precipitant solvent [25]. Then Nd:YAG transparent ceramics with fine microstructure and with the in-line transmittance of 83.8% at 1064 nm were obtained by the co-precipitated Nd:YAG raw powder and the optimized sintering schedule, combined with 0.14 wt% SiO_2 as sintering aid [26]. Nd:YAG nanopowders with the primary particle size of about 106 nm and low agglomeration were obtained by Ma et al. [27] via a modified co-precipitation method. And Nd:YAG ceramics with the in-line transmittances of 77.8% at 400 nm and 83.9% at 1064 nm were prepared at 1780 °C for 20 h. With the maximum absorbed pump power of 17.6 W, laser output of 4.5 W was obtained with a slope efficiency of 25.6% [27].

Well known vacuum sintering is the most widely used method to fabricate optical YAG ceramics. However, some defects, such as residual pores, can be hardly eliminated. The spark plasma sintering (SPS) technique allows for full densification of ceramics at a lower processing temperature with significantly short treatment durations, partly due to the high heating rates that are attainable comparing to other fabrication methods [28]. The use of SPS for the fabrication YAG has also been examined over the past years [29–31]. Nd:YAG transparent ceramics with the in-line transmittance of about 80% at the wavelength of 1000 nm were fabricated by Frage et al. using nano-sized powders and LiF additive [31]. However the optical transmission of the SPS-sintered Nd:YAG ceramics is still lower than the theoretical value due to the existence of residual pores. On the other hand, hot isostatic pressing has been shown to have a potential to remove the residual pores entirely [32–34], but it can bring another problems, such as contamination of the material with argon ions and gas inflated pores. Thus till now, there are few reports on the fabrication of YAG transparent ceramics using HIP post-treatment from YAG powders synthesized by the co-precipitation method [34]. And there is still space to optimize the fabrication process and to improve the optical quality of Nd:YAG transparent ceramics by this method.

In our previous work, Nd:YAG ceramics with the in-line transmittance over 80% at the near infrared wavelength were vacuum sintered at 1750 °C for 20 h from nanopowders synthesized by the co-precipitation method [35]. What's more, well-dispersed Nd:YAG powders with the particle size of about 100 nm were synthesized by co-precipitation method using MgO as the dopant and Nd:YAG transparent ceramics with the transmittance of 82.6% at 1064 nm were prepared by vacuum sintering at 1750 °C for 20 h [36]. In this work, Nd:YAG nano-powder were synthesized by the reverse co-precipitation method, and transparent Nd:YAG ceramics were obtained by vacuum sintering and successive HIP post-treatment. Effects of calcination temperature on phase evolution of the precursors and the sinterability of the Nd:YAG nanopowders were investigated. The optical transparency and the microstructure of the obtained Nd:YAG ceramics before and after HIP treatment were also investigated.

2. Experimental

The aluminum nitrate and yttrium nitrate solutions were prepared by dissolving aluminum nitrate nonahydrate $Al(NO_3)_3{\cdot}9H_2O$ (Tianjin Fine Chemicals, China) and $Y(NO_3)_3{\cdot}6H_2O$ (Tianjin Fine Chemicals, China) in distilled water, and neodymium nitrate solution was obtained by dissolving Nd_2O_3 (Alfa Aesar, 99.99%) in HNO_3 aqueous solution (Sinopharm Chemical Reagent Co., Ltd, COM). The metal nitrate solutions were mixed according to the stoichiometric ratio of $Nd_{0.03}Y_{1.97}Al_5O_{12}$ and further adjusted the concentration of Al^{3+} to 0.30 M. 1.5 M solution of ammonium hydrogen carbonate (AHC) (Sinopharm Chemical Reagent Co., Ltd, 99.0%) was prepared as the precipitation solution. Ammonium sulfate (99.0%, Sinopharm Chemical Reagent Co., Ltd.) was added into the precipitant solution as the dispersant. Chemical precipitation was performed by the reverse-strike method (adding salt solution to the precipitant) at the room temperature of about 25 °C. After aging for 2 h, and then being washed with distilled water and the ethanol for three times respectively, dried at 70 °C for 24 h, sieved through a 200-mesh screen in sequence, the precursors were obtained. The precursors were calcined at different temperatures for 4 h. The powders calcined at 1100 °C were ball milled with high-purity (99.7%) alumina balls for 2 h in anhydrous ethanol used as dispersion medium. 0.5 wt% TEOS was added as a sintering aid. The obtained slurry was dried, sieved through 200-mesh screen and then calcined at 800 °C for 1 h to remove water and residual organic matter. The as-prepared powders were uniaxially pressed in a 20 mm die at 50 MPa followed by CIP at 250 MPa. The green bodies were sintered at 1500–1800 °C for 10 h in vacuum and further HIPed at 1600 °C for 3 h in 200 MPa of argon.

Differential thermal and thermogravimetry analysis (DTA-TG) of the precursors was performed on a DTA-TG analyzer (Model STA 449 C, Netzsch, Germany). The heating rate was 10 °C/min up to 1200 °C under flowing air (20 mL/min). Chemical analysis was used to confirm the chemical stoichiometry of the solution to be equal: (Nd+Y):Al=3:5. X-ray diffractometry (XRD) device (Model D/max2200PC, Rigaku, Japan) was used to identify the phase of the as-calcined powders. Nickel-filtered Cu-K_α radiation and a scanning speed of 5 °C/min ranged in 10–80° (2θ) were used. The crystallite size of the YAG particles was determined in accordance with the Scherrer equation:

$$D_{XRD} = 0.89\lambda/\beta \cos\theta \tag{1}$$

where λ is the wavelength of Cu $K_{\alpha1}$ radiation (λ=0.1542 nm), β is the full-width at half-maximum (FWHM) of a diffraction peak at Bragg angle θ. Specific surface area analysis (Brunauer–Emmett–Teller analysis, BET) was performed by Norcross ASAP 2010 micromeritics with N_2 as the absorbate gas at 77 K. If the particles are assumed to be spherical, the average diameter of spherical particles can be calculated in accordance with the equation:

$$D_{BET} = \frac{6}{\rho{\cdot}S_{sp}}, \tag{2}$$

where S_{sp} – specific surface area of the powder, ρ – density of the material. Theoretical density of Nd:YAG is 4.55 g/cm^3. Microstructures of the powders and the obtained ceramics were observed with a field emission scanning electron microscope (FESEM, SU8220, Hitachi, Japan). The mean particle size was estimated by statistically analyzing 100 particles in the SEM photographs. The in-line transmittance of the sintered ceramics was measured with a UV–vis–NIR spectrophotometer (Model Cary-5000, Varian, USA). Grain sizes of the sintered samples were measured by the liner intercept method (more than 400 grain counted) from the scanning electron microscopy images of the thermally etched ceramics. The average grain size was determined by the multiplying the average linear intercept distance by a factor of 1.56.

3. Results and discussion

XRD patterns of the precursors and the powders calcined at different temperatures between 900 °C and 1300 °C for 4 h are shown in Fig. 1. The spectrum revealed the co-precipitated precursor (Fig. 1a) to be amorphous. Hexagonal YAP (JCPDS 16-219) and cubic YAG (JCPDS 82-0575) phases were detected at 900 °C. With further increase of the calcination temperature, the YAP phase disappears and YAG phase becomes the sole one. The precursor turned into pure YAG at 1000 °C without any traceable impure phases. Table 1 shows the crystallite size (D_{XRD}) of nanopowders calcined at different temperatures for 4 h. It can be seen that crystallite size increases with the temperature increased.

Fig. 2 shows the FTIR spectra of the precursor and the powders calcined at different temperatures for 4 h. Since nanopowders have a