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Fabrication and properties of Y_2O_3 transparent ceramic by sintering aid combinations

Dongyue Yan [a,b], Xiaodong Xu [a,b], Hao Lu [a,b], Yuwei Wang [a,b], Peng Liu [a,b,*], Jian Zhang [a,b,*]

[a] Jiangsu Key Laboratories of Advanced Laser Materials and Devices, School of Physics and Electronics Engineering, Jiangsu Normal University, Xuzhou, 221116 China

[b] Jiangsu Collaborative Innovation Center of Advanced Laser Technology and Emerging Industry, Jiangsu Normal University, Xuzhou, 221116 China

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ABSTRACT

Transparent Y_2O_3 ceramics were fabricated by the solid-state reaction and vacuum sintering method using La_2O_3, ZrO_2 and Al_2O_3 as sintering aids. The microstructure of the Y_2O_3 ceramics sintered from 1550 °C to 1800 °C for 8 h were analyzed by SEM. The sintering process of the Y_2O_3 transparent ceramics was optimized. The results showed that when the samples were sintered at 1800 °C for 8 h under vacuum, the average grain sizes of the ceramics were about 3.5 μm. Furthermore, the transmittance of Y_2O_3 ceramic sintered at 1800 °C for 8 h was 82.1% at the wavelength around the 1100 nm (1 mm thickness), which was close to its theoretical value. Moreover, the refractive index of the Y_2O_3 transparent ceramic in the temperature range from 30 °C to 400 °C were measured by the spectroscopic ellipsometry method.

1. Introduction

Due to its excellent chemical stability, low emission and small absorption coefficient in the IR region at high temperature, the Y_2O_3 transparent ceramics were used as excellent IR-windows materials [1,2]. In addition, compared with the conventional transmitting infrared window materials such as sapphire, AlON and $MgAl_2O_4$, Y_2O_3 ceramic also has longer cutoff wavelength and broad transparency range which is extremely important for IR-window applications [1–4].

For the window materials, the flexural strength of the materials is also very important as well as the transmittance of the materials. However, the flexural strength of the Y_2O_3 transparent ceramics is not very high at present [3,4]. According to the Hall-Petch relations, the ceramic with the smaller average grain size owns the higher flexural strength [5]. The fine grain size of the ceramics can be achieved by many approaches, such as using the nanocrystalline powder as the starting materials, two-step sintering method to enhance the densification with the minimum grain growth, and applying the sintering aids to suppress grain growth and etc [6–11]. In recent years, many sintering aids have been developed to improve the optical and flexural properties of the Y_2O_3 transparent ceramics [11–13]. The ThO_2 and HfO_2 were used as sintering aids in Y_2O_3 system, but they were less applied at present because of the toxicity and high expense [10]. In addition, high sintering temperature (2050–2100 °C) is necessary to achieve the high optical quality. Afterwards, La_2O_3, ZrO_2 as well as their combinations began to be used as sintering aids to fabricate high optical quality transparent ceramics [11–14]. Yang et al. has reported that transparent Y_2O_3 ceramics with the in-line transmittance over 80% at 1–6 μm can be fabricated by using La_2O_3 as the sintering aids [11]. In their studies, high doping concentration of La_2O_3 up to 10 mol% were used. Qing Yi et al. [12] has reported that the Y_2O_3 transparent ceramics were fabricated by co-doping with La_2O_3 and ZrO_2 for the first time. The transmittance of the samples was 79.93% at the wavelength around 1100 nm, and the average grain size was about 10 μm. Although the highly transparent Y_2O_3 ceramics have been fabricated, the average grain size of the Y_2O_3 fabricated is still over 10 μm, which is not favor to obtain the higher flexural strength [15].

In this work, highly transparent Y_2O_3 ceramics were prepared by a solid-state reaction method with La_2O_3, ZrO_2 and Al_2O_3 as composite additives. The microstructural evolution and optical transmittance of the Y_2O_3 samples which were vacuum sintered at different temperature were investigated.

2. Experimental procedure

Commercial high-purity Y_2O_3 (5 N, Jiahua Corp. Ltd., China) powders were used as the starting materials. High-purity La_2O_3 (4 N, Jiahua Corp. Ltd., China), ZrO_2 (99.5%, metal base, Alfa-Aesar, UK) and Al_2O_3 (4 N, Sumitomo Chemicals, Japan) powders were

* Corresponding author at: Jiangsu Key Laboratories of Advanced Laser Materials and Devices, School of Physics and Electronics Engineering, Jiangsu Normal University, Xuzhou 221116, China.

E-mail addresses: liupeng_tju@126.com (P. Liu), jzhang@jsnu.edu.cn (J. Zhang).

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used as sintering aids, and the doping concentration were fixed at 0.5 wt%, 3.0 wt% and 0.004 wt%, respectively. The weighted powders were mixed with ethanol by planetary ball-milled for 15 h with zirconia balls (3 mm, Tosoh, Japan). After dried at 55 °C for 24 h, the homogeneous powders were obtained. Then the powders were sieved through a 60 mesh screen. These homogeneous powders were calcined at 800 °C for 6 h and dry pressed into green bodies with a 16 mm stainless steel mold at 10 MPa. After that, the green bodies were cold isostatic pressed at 200 MPa. Eventually, green bodies were sintered at different temperature from 1550 to 1800 °C for 8 h under high vacuum degree (below 10^{-3} Pa). Then, the sintered samples were annealed at 1350 °C for 10 h in air. At last, the transparent Y_2O_3 ceramics were cut and polished, and samples with thickness of 1 mm were obtained. In order to obtain the flexural strength by three point bending tests, the samples were processed into the rectangular bars with the size of 3 mm*4 mm*36 mm. In addition, the surfaces of the rectangular bars were mirror polished and the edges of the bars were chamfered into 45°.

The morphology of the raw powders, the mixture powders after ball milling, the thermal etching surface and the fracture surface of the Y_2O_3 ceramics were recorded on a scanning electron microscope (SEM, JSM- 6510, JEOL, Kariya, Japan). The optical transmittance of Y_2O_3 ceramics was obtained by a UV–VIS–NIR spectrophotometer (Lambda 950, Perkin-Elmer, Waltham, MA) and Fourier transform infrared spectroscopy (Tensor 27, BRUKER OPTIK GmbH, Ettlingen, Germany). The flexural strength was obtained by Instron-5566 universal material testing machine (Norwood, MA, American). Furthermore, their refractive index under different temperature were tested by spectroscopic ellipsometry method (IR-VASE, J.A. Woollam, Lincoln, NE). The average grain size was measured by averaging over 200 grains and using a mean linear method. The density of the bulk ceramics was tested by the Archimedes method.

3. Result and conclusion

Fig. 1(a) and (b) presents the SEM image of the starting Y_2O_3, Al_2O_3 powders respectively. Fig. 1(c) shows the morphology of powder mixtures with sintering aids after ball milling. The average particle sizes of original Y_2O_3 powders was 4–5 μm with heavily agglomeration. The Al_2O_3 powders were homogeneous distributed and the mean particle sizes was about 200 nm. However, it can be obviously observed that the agglomeration of the particles was easily crashed by ball milling process, which is shown in Fig. 1(c). After the ball milling, the fine particle size powder mixtures appeared.

Fig. 2 shows the sintering map of Y_2O_3 ceramics in the temperature range of 1550–1800 °C. In addition to the different sintering temperature, all the holding time at the different sintering temperature was kept at 8 h, constantly. The grain size and the relative density increased with temperature increasing in general. It can be clearly found that below the 1650 °C, the density of sintered Y_2O_3 body increased rapidly, with the very slow grain growth rate. When the sintering temperature further raised to 1700 °C, the density was closed to the theoretical density, and the grain size was only 1.8 μm. With the sintering temperature further increase, the rapid grain growth happened. As the sintering temperature reached 1800 °C, the average grain size was around 3.5 μm, which was much smaller than the results reported by Ning et al. so that flexural strength value can reach to 203.2 MPa. In their work, La_2O_3 and ZrO_2 were used as the sintering aids and the average grain size of the Y_2O_3 ceramic sintered at the similar conditions was 9.11 μm [16]. This phenomenon indicated that the inhibition of grain growth have been carried out during sintering process through doping with three additives. It has been reported that La^{3+} ions enhanced the mobility of grain boundary while the Zr^{4+} ions strongly suppressed the migration of grain boundary [12–14]. Compared with the reference 16, the content of the Zr^{4+} in the study is higher than that of in the reference, which can cause the reduction of the average grain sizes. According to the literature [17], the small amount of Al_2O_3 can react with Y_2O_3, which were forming the eutectic $Y_4Al_2O_9$ (YAM) phase during the sintering process. The presence of the eutectic phase resulted in a significant increase in ionic diffusion rate. Furthermore, the increasing of the ionic diffusion rate at the grain boundaries can contribute to the higher grain-boundary mobility. All of these will improve the densification rate of the Y_2O_3 during sintering process after Al_2O_3 added. Therefore, the Y_2O_3 ceramics can be reach the full dense under the lower sintering temperature. And the transmittance of the Y_2O_3 can reach 82.1% around 1100 nm when it was vacuum sintered just at 1800 °C.

The thermal etching surface of the as-prepared Y_2O_3 ceramics

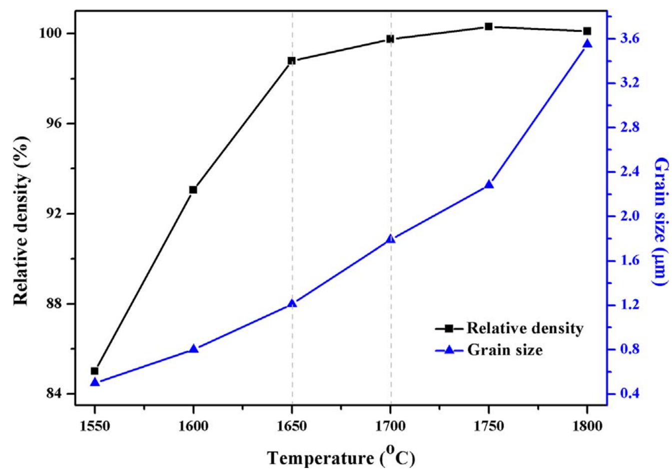

Fig. 2. Densification and grain growth behavior of the samples sintered at 1550–1800 °C for 8 h.

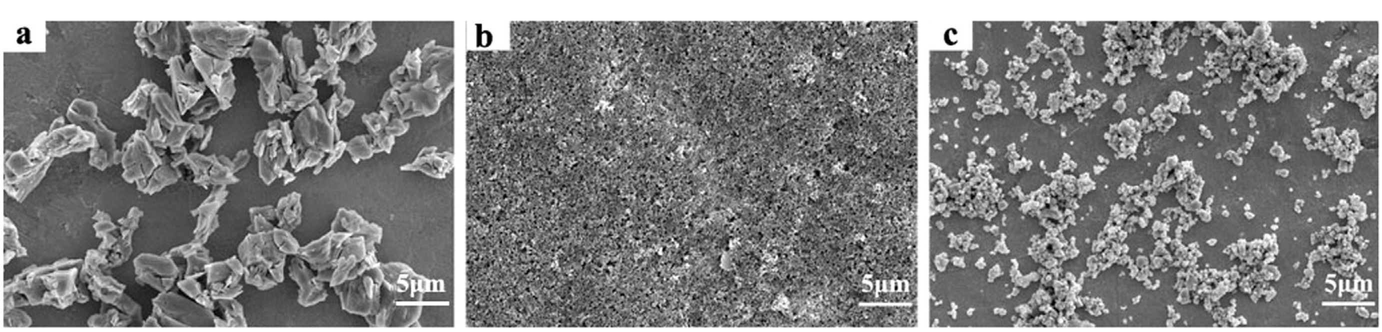

Fig. 1. SEM micrograph of the (a) raw powder of Y_2O_3; (b) raw powder of Al_2O_3; (c) powder mixture after ball-milling.

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