

Fabrication of Nd:YAG transparent ceramic with B₂O₃-SiO₂ as sintering aids

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Abstract. 1.0at% Nd:YAG polycrystalline ceramics were fabricated by solid-state reaction and vacuum sintering method using commercial Al₂O₃, Y₂O₃ and Nd₂O₃ as raw materials, SiO₂ and SiO₂ combined with B₂O₃ were used as additives. By changing the ratio of B₂O₃ to SiO₂, the effects of B₂O₃-SiO₂ doping on sintering kinetics and microstructure when sintered in vacuum have been investigated. The results indicate that the densification rate of the sample using only SiO₂ was lower than that of the ceramic samples using SiO₂ combined with B₂O₃ as additives. The transmittance spectra of the samples using compound additives decreased with the increasing B₂O₃, the optimal ratio of B₂O₃ to SiO₂ is B³⁺:Si⁴⁺=0.5, the maximum transmittance at 1064nm of the sample doped B₂O₃-SiO₂(B³⁺:Si⁴⁺=0.5) was 78.95%, nearly pore-free microstructure with the average grain size of 25μm was obtained in this highest transparent sample. The sample using B₂O₃-SiO₂ (B³⁺:Si⁴⁺=0.5) sintered to transparency at 1700°C which is lower than the sample using SiO₂ alone.

Introduction

Neodymium-doped yttrium aluminum garnet (Nd:YAG) has been considered to be one of the best solid-state laser materials in previous research for its good physical, mechanical and thermo-mechanical properties [1]. Many recent works have shown that the optical performance of Nd:YAG transparent ceramics is equivalent to or even better than that of Nd:YAG single crystals grown by the Czochralski method [2-3].

There have been many reports on the role and function mechanism of SiO₂ as sintering aid. The negative effects of SiO₂ have also been proved definitely that the less amount of SiO₂ affects the densification of the host material during processing but the more is not eliminated totally [4-5]. To decrease Si⁴⁺ content in Nd:YAG ceramics, we consider adding a sintering aid that interacts with the known liquid phase densification mechanisms during intermediate stage sintering of SiO₂ doped Nd:YAG, but which is then eliminated from the system. In other ceramic systems, SiO₂ co-doping with B₂O₃ has been shown to increase densification rates and lower sintering temperature[6].

In this paper, we report the synthesis of transparent Nd:YAG ceramics using B₂O₃-SiO₂ as a transient liquid phase sintering aid. By changing the ratio of B₂O₃ to SiO₂, we investigate the effects of B₂O₃-SiO₂ doping on sintering kinetics and microstructure when sintered in vacuum.

Experiment

The commercial high-purity Y₂O₃, Al₂O₃ and Nd₂O₃ powders were mixed according to the stoichiometric Nd_{0.03}Y_{2.97}Al₅O₁₂ with B₂O₃-SiO₂ as sintering aids. The B³⁺:Si⁴⁺ atomic ratio was varied between 0.5 and 2 while maintaining the total dopant content at 1.35mol%, which is equivalent to 0.14wt% SiO₂ doping. And ball milled in alcohol for (12-24) h. Then the mixture of powders was dried at 80°C for 8h and sieved through 100-mesh screen. After that, the powders were pressed into disks with 200MPa pressure. Subsequently, the Nd:YAG polycrystalline ceramics were obtained through

sintering under vacuum at (1600-1750)°C for 20h. Eventually, the specimens were annealed at 1450°C for 12h in air and mirror polished on both surfaces, which were shown in Table 1.

Tab1. shows the samples with different additives

Sample number	Additive types and content
Sample 1	1.35mol% SiO ₂
Sample 2	1.35mol% B ₂ O ₃
Sample 3	B ³⁺ :Si ⁴⁺ =0.5(total1.35mol%)
Sample 4	B ³⁺ :Si ⁴⁺ =1.0(total1.35mol%)
Sample 5	B ³⁺ :Si ⁴⁺ =2.0(total1.35mol%)

The crystal structures were indexed by X-ray diffractometer (XRD). The microstructure and the grain size of the samples were characterized by scanning electron microscope (SEM) and the optical transmittance spectra were measured using V-570 UV spectrophotometer.

Results and discussion

Fig.1 shows the XRD patterns of sample 1. It can be seen that the peaks of sample 1 can be well indexed as the cubic garnet structure of YAG and the same results were obtained in all the other samples. It indicates that, based on preliminary estimate, full transformation to YAG occurred during vacuum sintering despite the introduction of neodymium and additives.

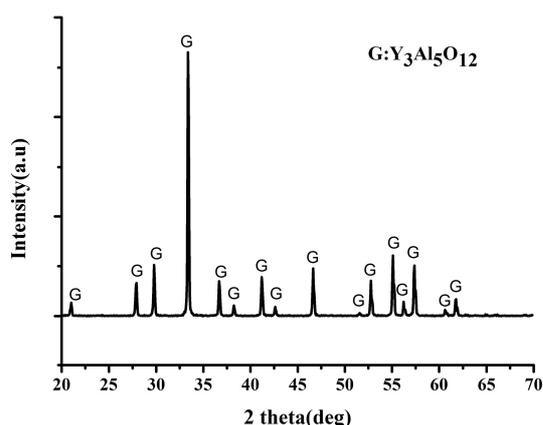


Figure1. XRD patterns of Nd:YAG ceramics samples 1 with 1.35mol% SiO₂

Fig.2 shows the optical transmittance spectra of the samples with different additives sintering at 1700°C for 20h, all the samples are 2.0mm thick. It can be seen that the transmittance of the sample 3, sample 4 and sample 5 decreases with the increasing B₂O₃. The transmittance of the sample 3 reaches 78.95%, 73.84% at 1064nm and 400nm respectively. The transmittance of sample 5 at 1064nm is only a little lower compared to sample 3 but decreasing sharply as the wavelength shifts toward the UV range, going down to only 60.95% at 400nm. Due to sintering at 1700°C, the transmittance of the sample 1 at 1064nm reaches 70.2%, which is lower than that of sample 3. Shown in Fig.3(a), there is still a few pore at the triple point and the average grain size is less than 50µm. For sample 2, B₂O₃ is known to vaporize from borosilicate glass melts at around 1500°C, it may act as a fugitive liquid phase sintering aid during intermediate stage sintering[7]. So there is no additive in sample 2 sintering at 1700°C, and the abnormal grain growth happened as confirmed by the observation of heterogeneous microstructures and the large diameter of the grains as shown in Fig.3(b).

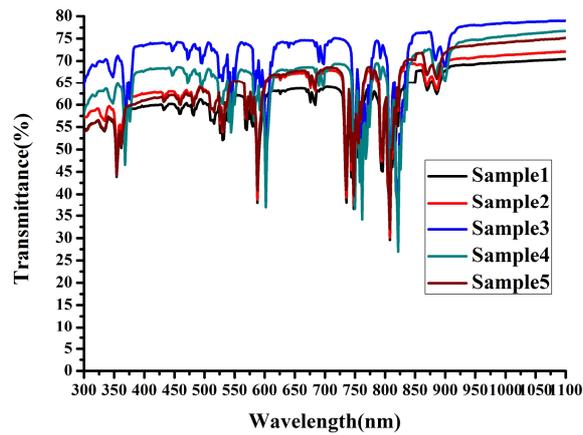


Figure 2. Optical transmittance spectra (300-1100nm) of Nd:YAG ceramics samples

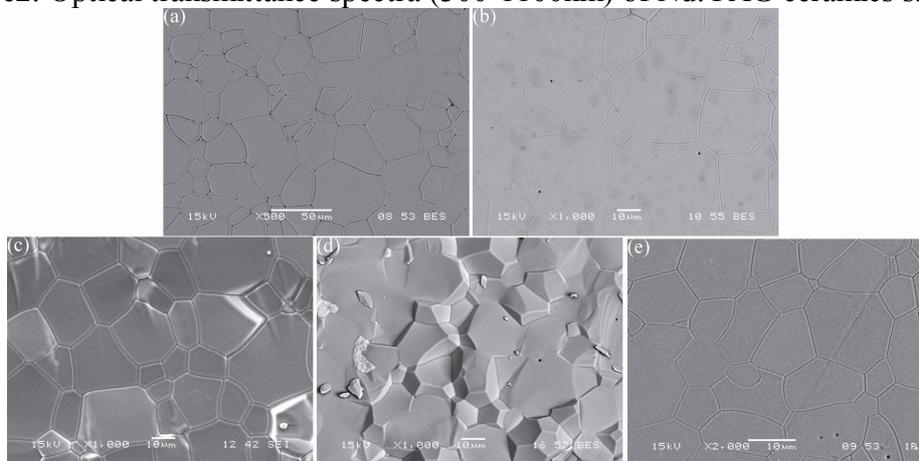


Figure 3. Microstructures of sample 1 (a), sample 2 (b), sample 3 (c), sample 4 (d) and sample 5 (e)

Fig. 3 shows that the SEM micrographs of the samples 1-5. It can be seen that the B_2O_3 - SiO_2 samples were nearly pore-free and the microstructures were qualitatively consistent. The grain size of the B_2O_3 - SiO_2 samples decreased with the increasing B_2O_3 from $25\mu m$ to $15\mu m$. For all temperatures, B_2O_3 - SiO_2 samples sintered to greater densities than sample 1 with only SiO_2 . The sintered density was highest in the sample 2 with $B^{3+}:Si^{4+}=0.5$ and decreased with increasing $B^{3+}:Si^{4+}$ atomic ratio shown in Fig. 4. It can be explained that increasing $B^{3+}:Si^{4+}$ ratio leads to decreased silica content after B^{3+} vaporizes. Densification and grain growth were shown to be critically dependent on SiO_2 content and increasing SiO_2 content in Nd:YAG ceramics increased both densification and coarsening[8]. Therefore, the sample 2 has the highest transmittance and the optimal ratio of B_2O_3 to SiO_2 is $B^{3+}:Si^{4+}=0.5$.

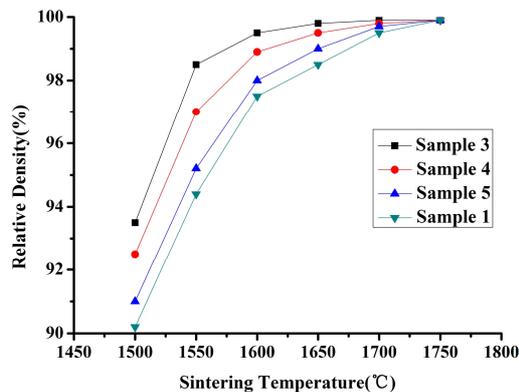


Figure 4. Relative density of samples sintered between $1500^{\circ}C$ and $1750^{\circ}C$

Fig.5 shows the transmittance spectra of sample 2 sintered at different temperature. It can be observed that the transmittance spectra of sample 2 sintered at 1750°C reaches 78.35%, 73.21% at 1064nm and 400nm respectively, which is only a little lower than that of the sample sintered at 1700°C. Therefore, the optimal sintering temperature of sample 2 was 1700°C. As shown in Fig.4, a full dense sample with $B^{3+}:Si^{4+}=0.5$ was sintered at 1700°C, which is about 50°C below the sintering temperature required to achieve transparency in only SiO_2 doped YAG ceramics.

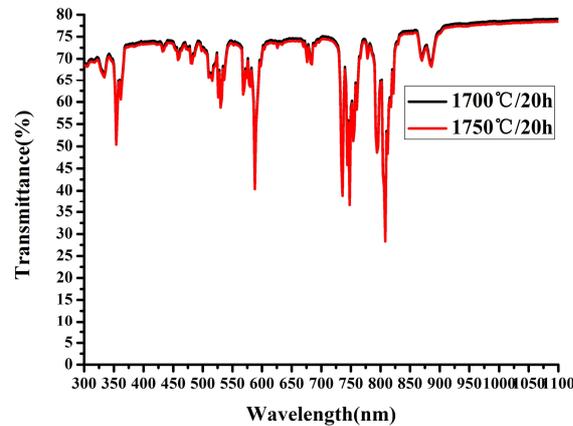


Figure5. Transmittance spectra of sample 2 sintered at different temperature

Conclusions

- (1) 1.0at% Nd:YAG highly transparent ceramics were successfully fabricated with B_2O_3 - SiO_2 as sintering aids by solid-state reaction and vacuum sintering method at 1700°C for 20h.
- (2) The optimal ratio of B_2O_3 to SiO_2 is $B^{3+}:Si^{4+}=0.5$. The transmittance of the sample 2 with $B^{3+}:Si^{4+}=0.5$ reaches 78.95%, 75.84% at 1064nm and 400nm respectively.
- (3) The highest transmittance, full dense and pore-free microstructure of sample with $B^{3+}:Si^{4+}=0.5$ were obtained at 1700°C for 20h, which is about 50°C below the sintering temperature in only SiO_2 doped YAG ceramics.

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