Synthesis and microstructure analysis of composite Nd:YAG/YAG transparent ceramics

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Transparent Nd:YAG/YAG composite ceramics are synthesized by solid-state reaction method using high-purity Y_2O_3, Al_2O_3, and Nd_2O_3 powders as raw materials. The mixed powder compacts are sintered at 1780 °C for 10 h under vacuum and annealed at 1450 °C for 20 h in air. The Nd:YAG/YAG ceramics exhibit a pore free structure with an average grain size of about 30 μm. The microstructure of the Nd:YAG/YAG composite transparent ceramics is studied and there is no interface between Nd:YAG and YAG ceramics. The Nd ion distribution in one grain is also studied, which shows that there is no segregation of Nd ions as in Nd:YAG crystals.

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Recent developments of laser diode (LD) pumped solid-state lasers have stimulated the development of high gain media[1−3]. Among the high gain media, the neodymium (Nd) doped yttrium aluminum garnet (YAG) is one of the most available laser materials. Nd:YAG crystals are widely used as the pumping media in solid-state lasers[4]. YAG crystallizes in the cubic system. Nd:YAG has the formula $\{Nd_xY_{1-x}\}_3Al_5O_{12}$, where Nd^{3+} substitutes for eight-coordinate Y^{3+}[5]. The ion radius of Y^{3+} is 0.101 nm and that of Nd^{3+} is 0.112 nm, exhibiting a difference of about 10.9%, and the segregation coefficient of Nd in YAG is about 0.18. So it is very hard to get highly doped and homogeneous Nd:YAG crystals.

Since 1995, polycrystalline ceramic laser materials have attracted much attention[6−9] because the optical quality has been improved greatly and highly efficient laser output can be obtained. The efficiencies are comparable or superior to those of single crystals. It is well-known that heat effect has bad impact on the laser efficiency and beam quality of high-power and large-energy solid-state lasers, while composite structure materials such as bonded crystal and composite ceramics could resolve the heat effect efficiently. Also it is easier for ceramics to get composite structures than crystals. As a result, the study on preparation and performance of composite structure materials has become a hot research topic.

In this letter, we report the results of the synthesis and microstructures of Nd:YAG/YAG composite transparent ceramics. Emphasis is laid on the Nd ion distributions in grains and boundaries.

High-purity powders of Al_2O_3, Y_2O_3, and Nd_2O_3 were used as starting materials. The starting powders were weighed respectively to achieve a chemical composition of 1 at.-% Nd:YAG and YAG, and mixed by ball-milling in anhydrous alcohol for 10 h, with an addition of 0.5 wt.-% tetraethyl orthosilicate (TEOS) as sintering aid separately. The mixtures were dried, sieved, and dry-pressed under 100 MPa into Φ20 mm disks, and finally one piece of Nd:YAG and one piece of YAG were cold-isostatically pressed under 250 MPa. The compacted Nd:YAG/YAG disks were sintered at 1780 °C under vacuum and then annealed at 1450 °C for 20 h in air. Mirror-polished samples (4 mm thick) on both surfaces were used to measure the optical transmittance and absorption spectra. The microstructure of Nd:YAG/YAG composite ceramics was studied using electron probe micro-analysis (EPMA) and energy dispersive analysis system of X-ray (EDX).

Figure 1 shows the quite transparent mirror-polished 1 at.-% Nd:YAG/YAG sample sintered at 1780 °C under vacuum and then annealed at 1450 °C for 20 h in air. The left photograph shows the face (the following microstructures that we analyze are all parallel to this face) perpendicular to the two polished round sides. The left side of the composite ceramics is undoped YAG which is white, while the other side is Nd:YAG and the color is a little bit red. From the characters behind the specimen (the right photograph), we can see that the sample is completely transparent.

Figure 2 shows the optical transmission spectrum of the Nd:YAG/YAG composite ceramics (thickness is 4 mm) from 400 to 1100 nm. The transmittance in the visible light region reaches 76% and it is about 79% near the 1100-nm region. For Nd:YAG/YAG composite ceramics, the absorption coefficients at 808 and 1064 nm are 3.82 and 0.18 cm^{-1}, respectively.

Figure 3 shows the EPMA microstructure of fracture surface of the specimen (the fracture surface is

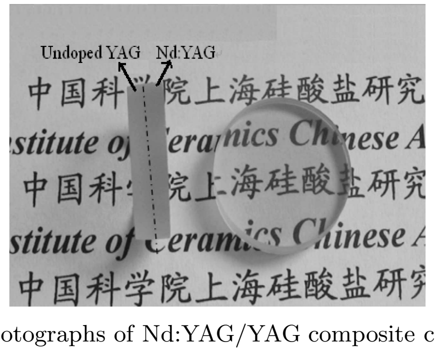

Fig. 1. Photographs of Nd:YAG/YAG composite ceramics.

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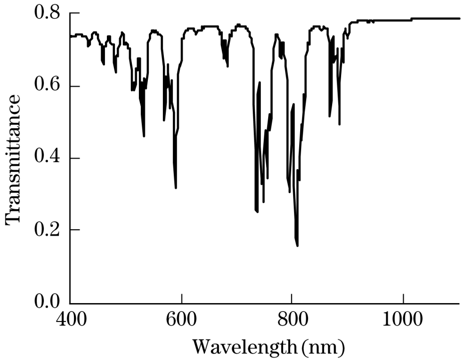

Fig. 2. Optical transmittance of the Nd:YAG/YAG composite transparent ceramics.

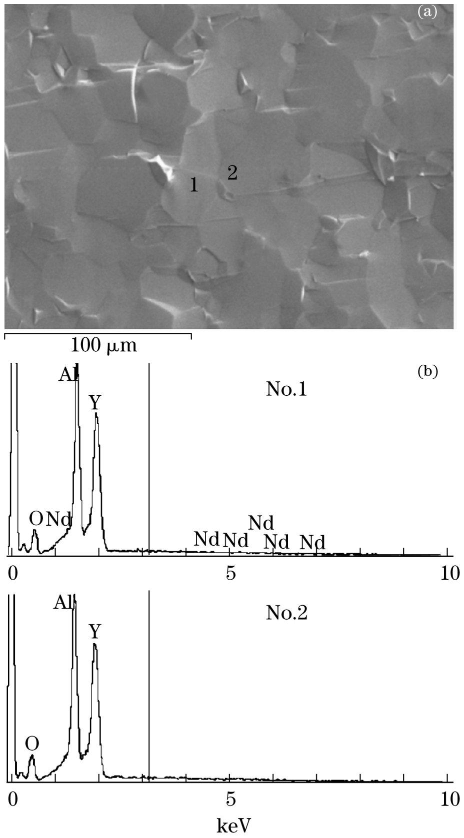

Fig. 3. (a) EPMA microstructure of fracture surface and (b) EDX spectra analysis of No. 1 and No. 2 grains.

perpendicular to the two polished sides) and EDX spectra analysis of No. 1 and No. 2 grains. The average grain size is about 30 μm and it appears relatively uniform. There are nearly no pores in or between the grains. Two grains were selected to make EDX spectra analysis. The results are shown in Table 1.

As we know, one side of the face is undoped YAG ceramics and the other side is Nd:YAG ceramics. So we first selected two grains a little bit far from the center and then moved to the center, at last we got the two grains close to each other, and one was undoped and the other was doped. According to the data from Table 1, we can see that there is no transition layer and no interface between Nd:YAG and YAG ceramics as in bonding crystals. This makes the properties of Nd:YAG/YAG composite ceramics superior to Nd:YAG/YAG bonding crystals.

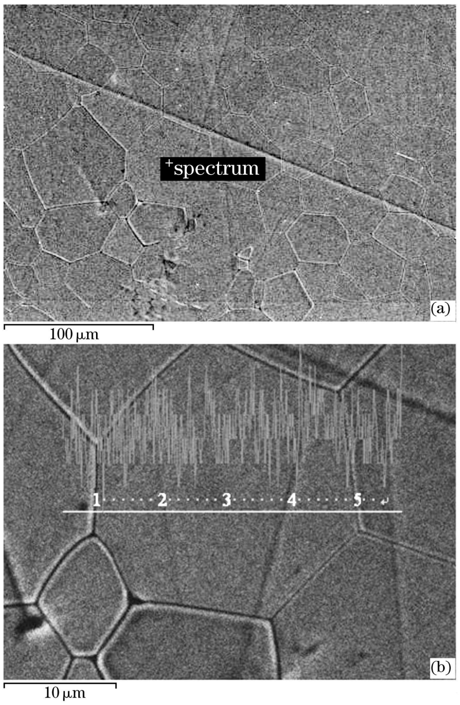

Fig. 4. (a) EPMA micrograph and (b) EDX spectra analysis of Nd:YAG grain of the mirror-polished and thermal etched surfaces of Nd:YAG/YAG composite transparent ceramics.

Table 1. Quantitative Analysis of No. 1 and No. 2 Grains by Energy Spectrum (at.-%)

	Y	Al	Nd	O
1	15.96±0.23	24.04±0.44	0	60
2	15.73±0.25	24.01±0.46	0.26±0.09	60

Figure 4(a) shows the EPMA micrographs of the mirror-polished and thermal etched surfaces of Nd:YAG/YAG composite transparent ceramics sintered at 1450 °C. It can be seen that there are little pores in or between the grains. From Fig. 4(b), one grain was selected to make EDX spectra analysis to get the Nd ion distribution. The results are shown in Table 2.

Table 2. Quantitative Analysis of Five Points of Nd:YAG Grain by Energy Spectrum (at.-%)

	Y	Al	Nd	O
1	16.16±0.28	23.51±0.50	0.27±0.09	60
2	16.25±0.28	23.49±0.50	0.26±0.09	60
3	15.60±0.28	24.02±0.50	0.38±0.09	60
4	16.12±0.28	23.69±0.50	0.19±0.10	60
5	15.87±0.28	23.87±0.50	0.26±0.10	60

From Table 2, we can see that the components of the five points in the grain are all very close to $Y_3Al_5O_{12}$. The Nd ion distribution in one grain is not so consentaneous and there is some nonuniformity (about 50% difference) in microcosmic area while the curve on the grains means the Nd distribution is uniform in macroscopic areas. There is no segregation as the phenomenon in Nd:YAG crystal growth, for which the Nd concentration is becoming higher during the melt solidification[10].

The causes of difference in one grain may be explained as follows. The diameters of Al_2O_3, Y_2O_3, and Nd_2O_3 raw materials are about hundreds of nanometers, and one ceramic grain needs about 100 Al_2O_3, Y_2O_3, and Nd_2O_3 powders. Because the Nd concentration is about 1 at.-%, it is impossible to make the Nd distribution uniform in microcosmic areas. During the sintering period, the Nd ions diffused and this made the Nd distribution tend to uniform. As for the grain in Fig. 4, maybe there was a Nd_2O_3 powder in point 3 and this caused the Nd concentration to be higher than the other areas.

In conclusion, Nd:YAG/YAG composite transparent ceramics were fabricated successfully by a solid-state reaction method. The Nd:YAG/YAG composite transparent ceramics exhibit a pore-free structure and the average grain size is about 30 μm. The transmittance of the Nd:YAG/YAG composite transparent ceramics reaches 79% at 1100 nm and 76% at 400 nm. The microstructure of the Nd:YAG/YAG composite transparent ceramics shows that there is no interface between Nd:YAG and YAG ceramics. The Nd concentration distribution is not uniform in microcosmic areas, while it is uniform in macroscopic areas.

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