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The physical properties of transparent $Y_3Al_5O_{12}$ Elastic modulus at high temperature and thermal conductivity at low temperature

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Abstract

We report experimental studies on the physical properties of transparent single and polycrystalline $Y_3Al_5O_{12}$ (YAG) used for laser materials or arc tubes for lamps. The elastic modulus and thermal conductivity of the YAG polycrystal were measured. The Young's modulus of the YAG polycrystal was 308 GPa at room temperature and decreased to 264 GPa at 1400 °C. Thermal conductivity increased with increasing grain size at temperatures below 150 K and was almost independent of grain size at higher temperatures. The thermal conductivity of the YAG polycrystal for a grain size of 7.5 μm was 1.1 W/cm K at 37 K, about seven times lower than that of the single crystal.

Keywords: B. Grain size; C. Thermal conductivity; YAG ceramics; Transparent ceramics; Elastic modulus; Phonon

1. Introduction

Transparent yttrium aluminum garnet ($Y_3Al_5O_{12}$:YAG) ceramic is an attractive material for optical and high-temperature structural applications [1,2]. The crystal structure of YAG is cubic, and optical transmission is about 84%. Therefore, the YAG ceramic is suitable as a tube material for lamps, because the optical property of transparent YAG ceramic is better than that of translucent alumina (Al_2O_3) ceramics, traditionally used for high-pressure sodium lamps or high-intensity discharge (HID) lamps. The fabrication methods and optical properties of transparent polycrystalline YAG ceramic were reported [3–6]. Now, these lamps used with a YAG polycrystalline arc-tube are being developed for many applications.

Although a YAG single crystal is one of many typical laser materials, it was thought that it was impossible to use the YAG polycrystal for laser materials. At the outset, their optical quality was very poor and it became essential to reduce the scattering sources in order to improve high optical quality. A neodymium doped-YAG (Nd:YAG) polycrystalline laser material produced by traditional method was first reported by Ikesue et al. [7]. This type of polycrystalline YAG ceramic was fabricated using a solid-state reaction of Y_2O_3 and Al_2O_3 by vacuum sintering. Laser operation was obtained with a slope efficiency of 28% under an end-pumping scheme using a 600-mW laser diode. However, the laser conversion efficiency of the Nd:YAG ceramic laser was much lower than that of commercially available high-quality single crystal lasers due to the high scattering losses.

Recently, we have been able to fabricate highly transparent solid-state laser polycrystalline YAG ceramics by the vacuum sintering technique and nanocrystalline technology [8]. Laser diode end-pumped ceramic Nd:YAG, with laser efficiency comparable to the best available single crystal Nd:YAG lasers, was developed in 2000 and 2001 [9,10]. For side-pumped high-power polycrystalline Nd:YAG rod lasers, the laser output power of 110 W was obtained with a slope efficiency of about 41% in 2004 [11]. The optical properties of our transparent YAG polycrystals were reported [9,12–14], while the laser damage threshold of YAG polycrystal was reported in [15].

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The mechanical properties and the thermal conductivity of transparent YAG ceramic are very important factors for the design and practical industrial used. Microhardness and fracture toughness of our YAG polycrystal were measured by Kaminskii et al. [16]. Hardness of the YAG polycrystal was found to be 10–15% higher than that of the YAG single crystal, and the fracture toughness was found to be least three times higher than that of the YAG single crystal. In [16], because no measurement of the elastic modulus of ceramic YAG was available, elastic modulus data of single crystal YAG were used to evaluate the fracture toughness of the YAG polycrystal. In this paper, we pay special attention to the elastic properties of YAG at high temperature because, for lamp applications, temperature of an arc tube wall exceeds 1000 °C.

On the other hand, thermal conductivity of our polycrystalline YAG was measured at room temperature and compared to that of the single crystal. The thermal conductivities of the Nd:YAG polycrystal and single crystal, were 10.5 and 10.7 W/mK, respectively, so no significant difference was found [17]. This result indicates that the quality of our polycrystalline YAG is high enough that the phonon–phonon interaction largely dominates phonon scattering by defect or impurities. However, a reduction of the thermal conductivity of the polycrystal can be observed compared to the single crystal at lower temperature mainly because of scattering at grain boundaries [18]. At low temperatures thermal conductivity of the polycrystalline YAG ceramic was measured with only two samples, i.e. transparent, or non-transparent [18].

Recently, cryogenic operation (close to liquid nitrogen temperature) of Ytterbium-doped YAG (Yb:YAG) is considered as a promising active material for the laser-driven fusion driver. Therefore, the particular measurement of thermal conductivity of ceramic is very important, not only for industrial engineering applications, but also for a scientific interest.

In this paper, the elastic (Young's) modulus of transparent YAG polycrystal was measured at high temperature. Moreover, thermal conductivity was measured at low temperature with three transparent polycrystalline samples with different grain size.

2. Fabrication of the transparent polycrystalline YAG ceramic

The transparent polycrystalline YAG sample was fabricated by slip casting and vacuum sintering method [8]. Aqueous solutions of aluminum, and yttrium chlorides were mixed together. The mixed aqueous solution was added dropwise and mixed with aqueous solution of ammonium hydrogen carbonate. Then steps of filtration and washing with water were repeated several times, and the obtained powder material was dried for 2 days at 120 °C in an oven. The obtained precursor consists of yttrium, aluminum hydrate ($Y(OH)_3$, $Al(OH)_3$) and those carbonate ($Y(CO_3)_3$, $Al(CO_3)_3$). This precursor was calcinated at approximately 1200 °C to produce raw YAG oxide powder with an average particle diameter of about 200 nm. The powder was ball-milled with solvent, binder

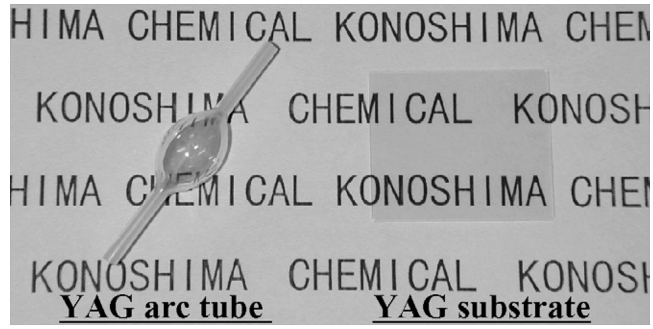

Fig. 1. Transparent YAG ceramics (an arc tube and a substrate).

and dispersion medium, and mixed for 24 h. The milled slurry was put into a gypsum mold and dried to obtain a desired form. By slip casting forming method, high density and uniform green body was obtained. After removing the organic components by calcinations, the materials were sintered in vacuum at 1650–1750 °C for several hours. All of the resulting samples were transparent as shown in Fig. 1.

Fig. 2 shows the microstructure of polycrystalline YAG sample, which was sintered at 1700 °C for 10 h. The microstructure was measured by using a scanning electron microscope (SEM). The microstructure of the boundary area was observed on a thermally etched (1400 °C × 2 h, in air) sample. The SEM image shows that the grain size of YAG polycrystal is very small and uniform. Moreover, it was clear that the scattering source (pore, second phase, etc.) did not exist.

The grain size was determined with a SEM by freely drawing a line on a high resolution image of the sample. When the length of the line was C, the number of grains on this line was N, and the magnification was MAG, the average grain size was given by next equation [19].

$$\text{The average grain size} = \frac{1.56 \times C}{N \times \text{MAG}} \quad (1)$$

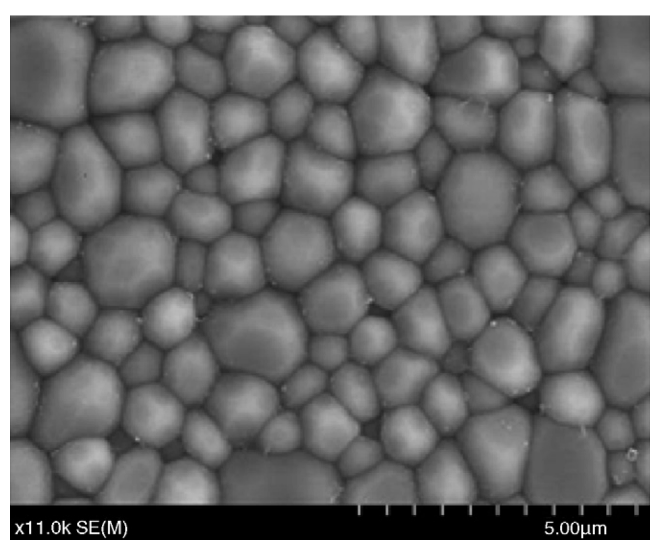

Fig. 2. The SEM image of a microstructure of the YAG polycrystal, sintered at 1700 °C for 10 h.

3. Results and discussion

3.1. Elastic modulus

The Young's modulus of polycrystalline sample was measured with the resonance methods in accordance with the Japan Industrial Standard (JIS) R1605 and used with elastic modulus testing machine (Model: DEM-11H, Kyoto-Densi-kougou, Japan). The length (L_t), width (W), and thickness (t) of measured sample were 100, 25, and 2 mm, respectively. The sample was suspended from two alumina fibers inside the furnace. The resonant frequency (f) was measured at each temperature. The young's modulus (E) was calculated from

$$E = 0.9465 \times 10^{-6}\frac{M f_2}{W} \times \left(\frac{L_t}{t}\right)^3 \times \left\{1 + 6.59\left(\frac{t}{L_t}\right)^2\right\} \quad (2)$$

where M is the weight of measured sample. Temperature dependence of the Young's modulus is shown in Table 1. Table 1 also shows the resonant frequency and the Poisson's ratio at each temperature. The Young's modulus of ceramic was found to be 308 GPa at room temperature to decrease to 264 GPa at 1400 °C.

The mechanical properties of transparent polycrystalline YAG were reported in [20], and the Young's modulus of their ceramic was about 290 GPa, at room temperature. Moreover, the Young's modulus of a YAG single crystal is 283 GPa at room temperature [20]. Those two values are almost the same. However, the Young's modulus of our YAG polycrystal is clearly higher than those literature values. The Young's modulus of polycrystalline ceramic generally depends on the porosity of sample [21]. The porosity of our ceramic is very low. In the case of single crystal, theoretical values were different at each axial direction. So we think that the difference between the two values is caused by those reasons.

3.2. Thermal conductivity at low temperature

Thermal conductivity was measured using a steady heat flow method at low temperature [18], and this system consists of two thermometers, a sample holder, a heater holder and a vacuum can. The measurement method is as follows. Heat from the heater flows through the sample and the thermal conductivity is calculated from the heater power (Q) and the different in temperature (ΔT) between two thermometers set on the samples. Then the thermal conductivity λ is obtained by

$$\lambda = \frac{Q}{S}\frac{\Delta x}{\Delta T} \quad (3)$$

where Δx is the distance between the thermometers and S the cross sectional area of the sample. The sample holder contains the sample, which is 2 mm × 2 mm in cross section and 10 mm in length. A hermetic ceramics packages were used for the thermometers and they were firmly attached to the holders. The thermometer holders are clamped to the sample by the sharp edges. Manganese wire was wound onto the heater holder as a heat source, and phosphor bronze wire was used as signal leads. All wires attached to the heat sink thermal anchor were maintained at a constant temperature by the controller. A gas flow type refrigerator was used to control the temperature of heat sink. More details about this measurement technique can be found in [18].

Thermal conductivity was measured at low temperature region from 5 to 200 K. Three transparent polycrystalline YAG samples were prepared for measurement, and each sample had different average grain size: 3, 4, and 7.5 μm, respectively. For comparison, thermal conductivity of a YAG single crystal was also measured.

Fig. 3 shows the experiment results on the thermal conductivity of the single crystal and polycrystal samples. Above 100 K, the thermal conductivities of the tested sample was about the same. In fact, thermal conductivity of the polycrystal and single crystal is the same from room temperature down to about 150 K. However, differences among the samples become apparent below 100 K. At 25 K, thermal conductivity of the single crystal was largest, and it was about 8 W/cm K. On the other hand, the thermal conductivity of polycrystal was about seven times lower than that of the single crystal. The maximum thermal conductivity of polycrystal samples, which had 3, 4, and 7.5 μm grain size, was 0.64, 0.99,

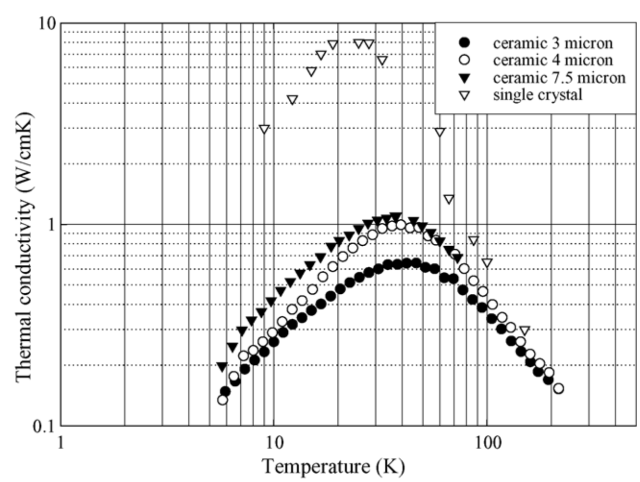

Fig. 3. The experiment results on the thermal conductivity of the YAG single crystal and polycrystal.

Table 1
Temperature dependence of the Young's modulus and the Poison's ratio

Temperature (°C)	R.T (25)	200	400	600	800	1000	1200	1400
Resonant frequency (Hz)	1678	1666	1654	1634	1618	1602	1580	1552
Young's modulus (GPa)	308	304	300	293	287	281	274	264
Poison's ratio	0.233	0.238	0.234	0.224	0.228	0.216	0.221	0.23

and 1.1 W/cm K, at temperature of 46, 39, and 37 K, respectively. Of course, the grain size of single crystal was very large. So, it can be seen that the temperature which had highest thermal conductivity, fall down according to increasing the grain size. Therefore, the grain boundary of a polycrystal contributed to the thermal conductivity of the ceramics samples, at very low temperature.

At low temperature, the polycrystal has lower thermal conductivity than that of a single crystal, because the phonon mean free path becomes longer and phonon scattering occurs at the grain surface in the ceramic, which consists of strongly bound fine single crystals.

4. Conclusions

We measured the Young's modulus and thermal conductivity of the transparent polycrystalline YAG ceramic. The Young's modulus of polycrystalline YAG was 308 GPa at room temperature, and decreased to 264 GPa at 1400 °C. Estimated Young's modulus was higher than that of previous reported value. We think that the difference is caused by the difference of porosity or density of both ceramics.

At low temperature, thermal conductivity of polycrystal ceramics was dependent on the grain size, and higher thermal conductivity values were found for the samples with larger grain size. The maximum thermal conductivity of polycrystalline samples, which had 3, 4, and 7.5 μm grain size, was 0.64, 0.99, and 1.1 W/cm K, at temperature of 46, 39, and 37 K, respectively. Thermal conductivity of this polycrystal was about seven times lower than that of the single crystal, at low temperature.

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