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Fabrication of YAG transparent ceramics by two-step sintering

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Abstract

Yttrium aluminum garnet (YAG) nanopowders with mean particle size of about 50 nm synthesized by a modified co-precipitation method were used to sinter bulk YAG ceramic by two-step sintering method. Full densification was achieved by heating the sample up to 1800 °C followed by holding at 1550 °C for 10 h. Transparent YAG ceramics were obtained by suppressing grain-boundary migration while promoting grain-boundary diffusion during the two-step sintering process. The microstructure of the YAG ceramic is homogeneous without abnormal grain growth and the transmittance of the sintered sample is 43%.

Keywords: A. Grain growth; A. Sintering; B. Grain boundaries; B. Grain size

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1. Introduction

Recently there is a great interest in the fabrication of transparent yttrium aluminum garnet (YAG) ceramics, because the polycrystalline YAG ceramic is a promising substitute for single crystal YAG due to its excellent optical properties, high-temperature mechanical properties, and its far lower production cost than that of single crystalline YAG [1,2]. Fabrication of transparent YAG ceramics by traditional pressure-less sintering technique has been reported during the past decades [3–9]. Researchers have also used non-conventional sintering method such as spark plasma sintering to achieve dense YAG ceramics [10]. Preliminary studies on the synthesis of YAG nanopowders obtained by a modified co-precipitation method were reported by us. We have also shown that the nanocrystalline YAG particles can be sintered to transparency by vacuum sintering at 1700 °C for 5 h with tetraethyl orthosilicate (TEOS) as sintering additive [11].

Many reports on the fabrication of transparent YAG ceramic were conducted by prolonged sintering in vacuum at quite high temperatures, therefore abnormal grain growth could occur with ease. A two-step sintering technique for Y_2O_3 was first introduced by Chen and Wang [12,13], in which the sample was first heated to a higher temperature then cooled down to a lower temperature to suppress grain-boundary migration, and held at that temperature till full densification. Such a two-step sintering method is effective to obtain nanoceramics such as Si_3N_4 and Al_2O_3 nanoceramics [14,15]. In this paper, the two-step sintering (TSS) concept was adopted for the sintering of transparent YAG ceramics.

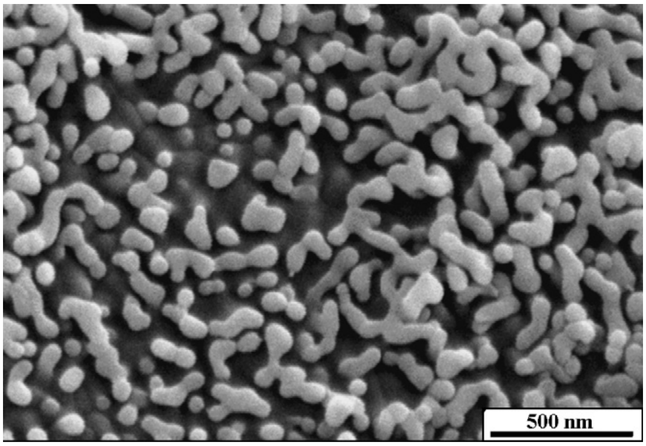

Fig. 1. SEM micrograph of YAG nanocrystalline calcined at 1100 °C.

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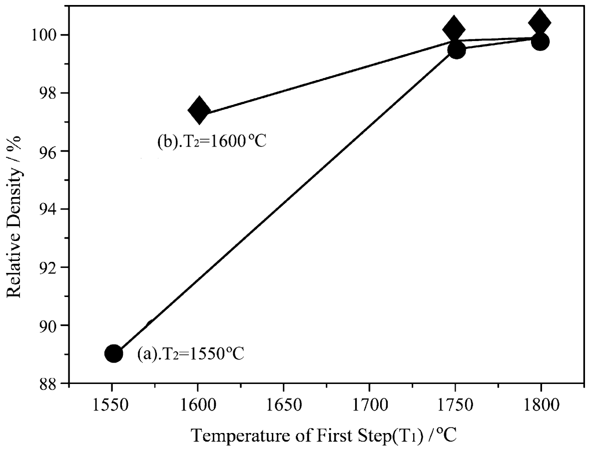

Fig. 2. Relative density of the samples in TSS as a function of peak temperature T_1: (a) $T_2 = 1550$ °C and (b) $T_2 = 1600$ °C.

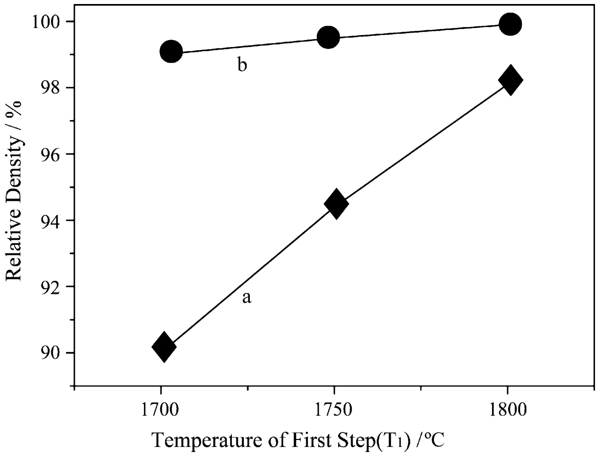

Fig. 3. Relative densities of samples (a) in single-step sintering at T_1 and (b) in the TSS process in which T_2 is 1550 °C.

2. Experimental procedure

The YAG nanopowders were synthesized by the modified co-precipitation method [10]. Fig. 1 shows the SEM morphologies of YAG particles calcined at 1100 °C which are composed of well-dispersed fine particles with particle size of 50 nm in average.

TEOS (0.5% mass) was used as a sintering additive for the sintering of YAG powders. The powder mixtures were milled in ethanol for 2 h, then dried and calcined at temperature up to 800 °C in air to remove residual organic materials. The calcined powders were cold pressed in a steel mold at a pressure of 10 MPa and then cold isostatic pressed at 200 MPa to 40–45% theoretical density. The compacts were sintered in vacuum furnace at 1500–1800 °C. The vacuum degree was 10^{-3} Pa during the sintering period. The compacts were rapidly heated to a peak temperature (named hereafter as T_1, $T_1 = 1700$, 1750, 1800 °C) without holding time, and then immediately cooled down to a lower temperature (named hereafter as T_2, $T_2 = 1500$, 1550, 1600 °C) and held at T_2 for 10 h. In order to establish the two-step sintering conditions, single-step sintering runs were conducted at temperature T_1 and T_2, respectively. In the

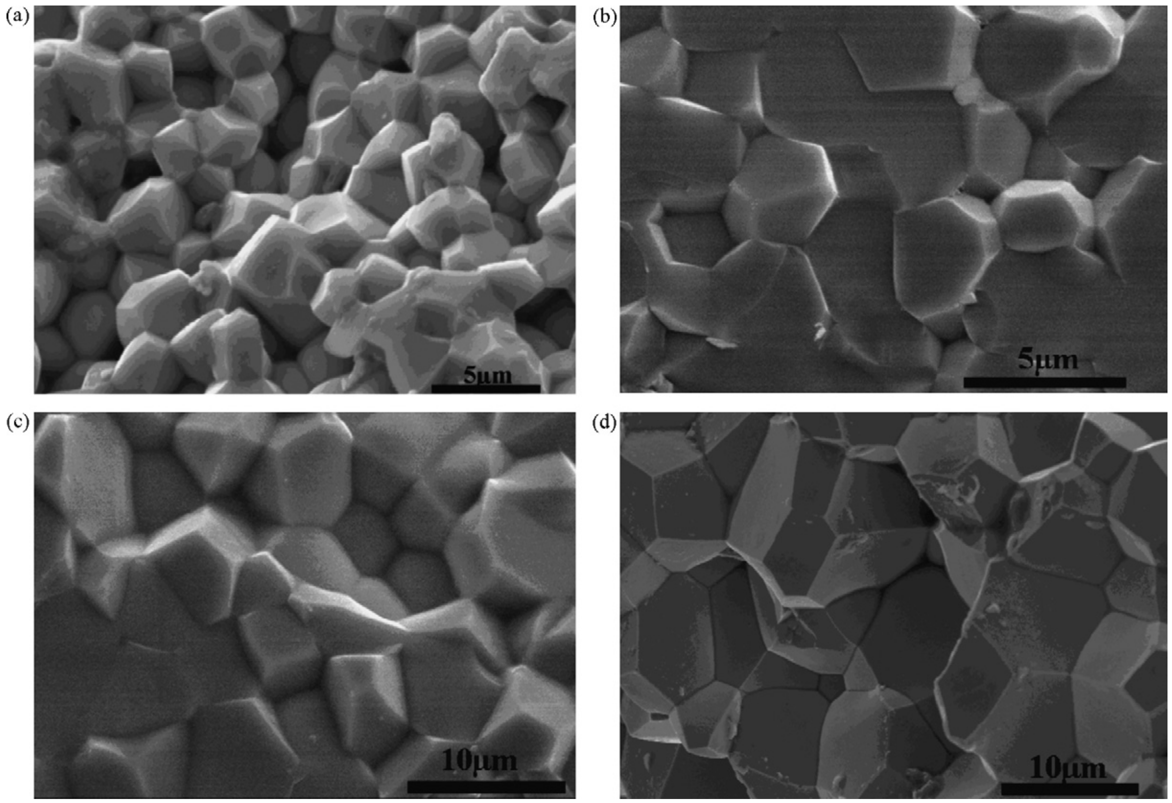

Fig. 4. Fracture surfaces of YAG samples sintered by the TSS process: (a) 1750–1550 °C; (b) 1750–1600 °C; (c) 1800–1550 °C; (d) 1800–1600 °C.

single-step sintering runs at temperature T_1, the samples were rapidly heated to T_1 and immediately cooled down to room temperature. In the single-step sintering runs at temperature T_2, the samples were heated to T_2 and held at T_2 for 10 h.

The fracture surfaces of sintered YAG specimens were observed using scanning electron microscopy (SEM, HITACHI S-4300). Specimens for transmittance measurement were cut into 1.0 mm thick slices and both surfaces were mirror polished. Transmittance of the transparent YAG ceramics was measured over the wavelength region from 200 to 800 nm using a UV spectrometer (Perkin-Elmer UV–Vis Tu-1901). The densities of the samples were measured using Archimedes method.

3. Results and discussion

The density and the grain size of the sintered YAG as a function of the peak temperature are shown in Table 1. In the single-step sintering runs at temperature of T_1, the maximum densities of the YAG ceramics reached 90.0–98.1%TD corresponding to the peak temperature of 1700–1800 °C. In the single-step sintering runs at temperature of T_2, The density of samples varied between 76.8%TD and 97.2%TD when sintered at 1500–1600 °C for 10 h. In the TSS process, the product densities of above 99.0%TD were generally achieved.

Fig. 2 shows that the relative density of the samples prepared by the TSS process at different holding temperatures of T_2 strongly depends on the peak temperature of T_1. It is impossible to achieve full densification merely by sintering the samples at temperatures below 1600 °C for 10 h. However, when an adequate peak temperature of T_1 was applied before holding the samples at temperature of T_2, over 99.9%TD were achieved at the same temperature of 1600 °C and the same holding time of 10 h. Fig. 3 shows the relative densities of the samples in the single-step sintering runs at T_1 and in the TSS process in which T_2 is 1550 °C. It is evident that when the two steps were conducted in succession, the density of the sample is higher than that in either of the single-step sintering runs. Therefore, the second step is effective to promote further densification. It is because a sufficiently high starting density is obtained at temperature of T_1, all pores in YAG become subcritical and unstable against shrinkage. Continuous framework formed

Table 1
Relative density and grain size for the specimens on different sintering conditions

Sintering condition	Designation of sintered samples	Temperature (°C)	Relative densities (%)	Grain size (μm)
Single-step sintering				
T_1-0 h	a	1700	90.0	2.11
	b	1750	94.4	2.86
	c	1800	98.1	5.33
T_2-10 h	d	1500	76.8	0.65
	e	1550	88.9	0.81
	f	1600	97.2	1.33
Two-step sintering				
(T_1-0 h) + (T_2-10 h)	g	1700–1550	99.0	2.62
	h	1750–1550	99.5	3.18
	i	1750–1600	99.8	3.56
	j	1800–1550	99.9	6.04
	k	1800–1600	99.9	8.36

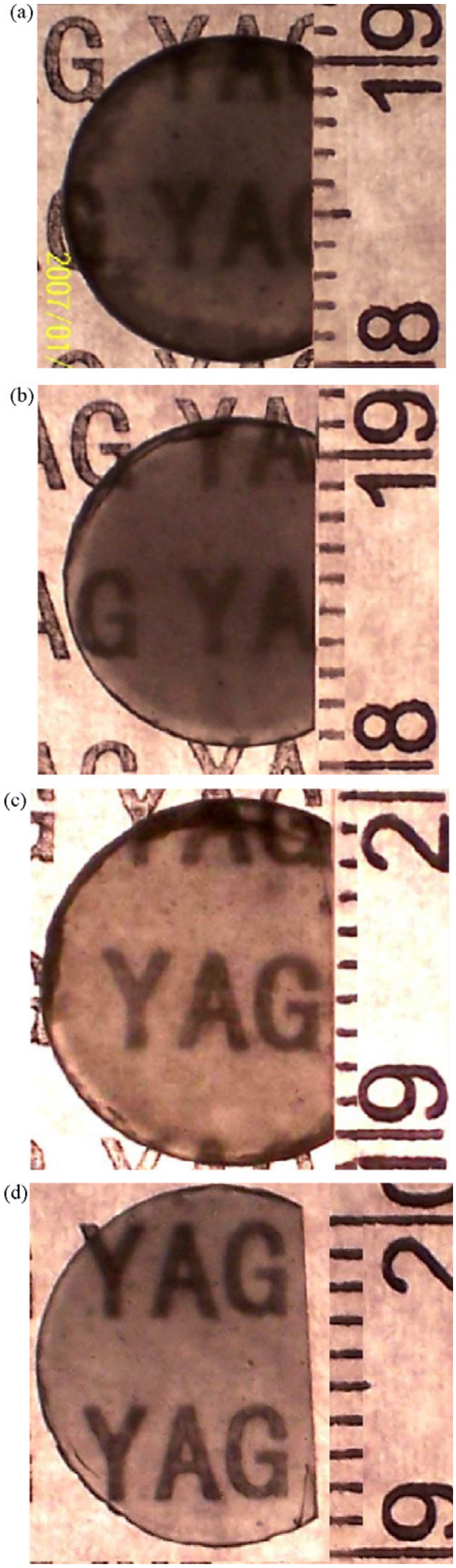

Fig. 5. The photographs of the mirror-polished YAG samples sintered by the TSS process.

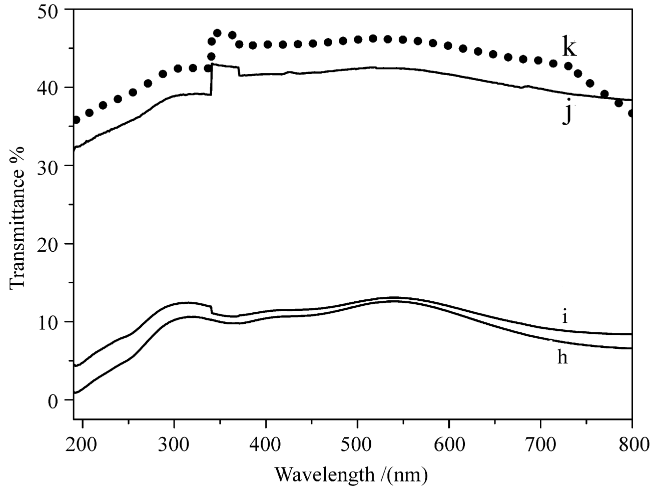

Fig. 6. The optical transmission spectrums of the sintered YAG samples h, i, j and k respectively.

among YAG grains and acted as atom diffusion path driving the subsequent sintering step. The activation energy for the grain-boundary diffusion is far lower than that for grain-boundary migration, thus, the energy obtained at temperature of T_1 is sufficient for the grain-boundary diffusion during the subsequent holding process at temperature of T_2. Promoted by grain-boundary controlled kinetics, full densification of the samples was achieved.

Fig. 4 is the fracture surfaces of the sintered YAG samples by the TSS process. The nearly pore-free microstructures of the samples remained homogeneous without abnormal grain growth. The grain size increases with the sintering temperature, the corresponding grain size of the samples h, i, j and k is 3.18, 3.56, 6.04 and 8.36 μm. The grain size of the samples h, i and j grow much less than that of the sample k.

Fig. 5 shows the mirror-polished YAG samples sintered by the TSS process with 11 mm in diameter and about 1.0 mm thick, which were all transparent or translucent. Fig. 6 shows the optical transmission spectrums of the sintered YAG samples. The transmittance of samples h, i, j and k is 10.5%, 12.6%, 43% and 47%, respectively. It increases with the increase of the value of both the first step sintering temperature and the holding time at the second step sintering temperature. When the sample being sintered at 1800 °C and followed by 1550 °C for 10 h, grain-boundary migration was effectively suppressed and transparent YAG ceramics was obtained with grain size being 6 μm. The temperature of 1550 °C may be the lowest pressure-less sintering temperature ever employed to obtain transparent YAG ceramics.

4. Conclusion

YAG transparent ceramics was prepared with 0.5 wt% TEOS as sintering additive using TTS process. The microstructures of the as fabricated YAG ceramics are homogeneous. The transmittance of YAG ceramic increases with the increase of the value of both the first step sintering temperature and the holding time at the second step sintering temperature. Up to 43% transmittance was obtained when the sample was sintered at 1800 °C as the first step and at 1550 °C for 10 h as the second sintering step, and the mean grain size of the sample is 6 μm.

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