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Effects of Shaping Conditions on the Microstructure and the Mechanical Property of the Al_2O_3-YAG Eutectic Composite Produced by Melting the Al_2O_3-YAP Eutectic Structure

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When the Al_2O_3-YAP metastable eutectic system was heated above the metastable eutectic temperature but below the equilibrium eutectic temperature, undercooled melt was formed. Solidification in the equilibrium path accompanied the melting of the metastable eutectic system, resulting in a fine and uniform eutectic structure. A novel solidification process using undercooled melt formation was developed. In this paper, the influences of the initial Al_2O_3-YAP eutectic particle size, forming pressure and holding time at the maximum temperature on the microstructure and/or the mechanical property of the Al_2O_3-YAG compact were examined to optimize the forming conditions. The Al_2O_3-YAG compact produced from the Al_2O_3-YAP eutectic particles with diameters less than 20 µm exhibited a higher flexural strength and the smaller porosity as compared to that produced from Al_2O_3-YAP eutectic particles with diameters ranging from 20 µm to 45 µm. The volume fraction of the porosity was approximately 4% under a forming pressure of 10 MPa and a holding time of 60 s. The flexural strength of the Al_2O_3-YAG compact decreased with the increase in the holding time due to the increase in the lamellar spacing. For Al_2O_3-YAP eutectic particles with diameters less than 20 µm, a low forming pressure of 10 MPa and a short holding time of 60 s were sufficient to synthesize a dense Al_2O_3-YAG compact. Thus, the solidification technique that uses undercooled melt produced by melting the Al_2O_3-YAP metastable eutectic system has good advantages for shaping. [doi:10.2320/matertrans.MB200705]

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1. Introduction

Unidirectionally solidified Al_2O_3-based eutectic ceramics have gained considerable attention as heat-resistance materials because of their superior mechanical properties at high temperatures.[1–4] In recent years, Waku *et al.* reported that single crystal Al_2O_3/single crystal YAG($Y_3Al_5O_{12}$: yttrium-aluminum-garnet) eutectic composites exhibited a flexural strength of 360–400 MPa at temperatures ranging from room temperature to 2073 K.[2–4] In addition to excellent mechanical properties at high temperatures, some interesting solidification phenomena have been reported in the Al_2O_3-Y_2O_3 system.[5–7] Caslavsky and Viechnickl discovered the two eutectic reactions by optical differential thermal analysis in the Al_2O_3-rich part of the Al_2O_3-Y_2O_3 phase diagram—one is the Al_2O_3-YAG equilibrium eutectic reaction at 2099 K and the other is the Al_2O_3-YAP metastable eutectic reaction at 1975 K, as shown in Fig. 1.[7]

The Al_2O_3-YAP metastable eutectic system can be selected in the composition range of 13.5–28.5 mol% Y_2O_3.[7–10] When the melt that has a metastable eutectic composition is heated to temperatures above 2273 K, the Al_2O_3-YAP metastable system is indispensably selected. In our previous work,[8–15] heating the Al_2O_3-YAP metastable eutectic system up to temperatures above the metastable eutectic temperature, undercooled melt for Al_2O_3-YAG equilibrium eutectic system was produced due to melting of the Al_2O_3-YAP metastable eutectic structure at the metastable eutectic temperature.

The solidification of the Al_2O_3-YAG equilibrium eutectic system accompanies the melting of the Al_2O_3-YAP metastable eutectic system. Thus, the endothermic heat generated due to melting is nearly compensated by the exothermic heat generated due to the solidification. As a result, the solidification rate becomes relatively high, resulting in a fine and uniform eutectic structure. On the basis of these findings, a novel solidification technique utilizing the undercooled melt shaping has been developed.[8–15]

Our previous paper demonstrated that Al_2O_3-YAG castings with a relatively complex shape were produced by the undercooled melt shaping.[13] For example, the groove shape of the molybdenum mold (300 µm in width and 1 mm in thickness) was well transferred to the castings. Al_2O_3-YAG eutectic particles with diameters in the range of 25–40 µm were used to produce the castings. Porosities of diameters in the range of 5–10 µm were observed in the top part of the fins. Since the Al_2O_3-YAG eutectic structure produced from undercooled melt could be influenced by the initial particle size of the Al_2O_3-YAP eutectic particles, the study of the influence of the initial particle size of the Al_2O_3-YAP on the Al_2O_3-YAG eutectic structure is of importance. In addition, it is necessary to optimize the casting conditions during the heating process in order to synthesize dense castings with a uniform microstructure.

In this paper, the influence of the initial Al_2O_3-YAP particle size, forming pressure and holding time at the maximum temperature on the Al_2O_3-YAG solidification structure and the flexural strength of Al_2O_3-YAG eutectic castings were examined in order to optimize the process conditions for undercooled melt shaping.

2. Experimental Procedure

α-Al_2O_3 powder (99.99%) and Y_2O_3 powder (99.99%) were weighed according to Al_2O_3-23.5 mol% Y_2O_3 which is an Al_2O_3-YAP metastable eutectic composition. These powders were ball milled with ethanol for 24 h to obtain homogeneous slurry that was then dried at 473 K. The mixed powder was calcined at 1573 K for 3 h in air. The calcined powder was melted in a Mo crucible by an induction furnace

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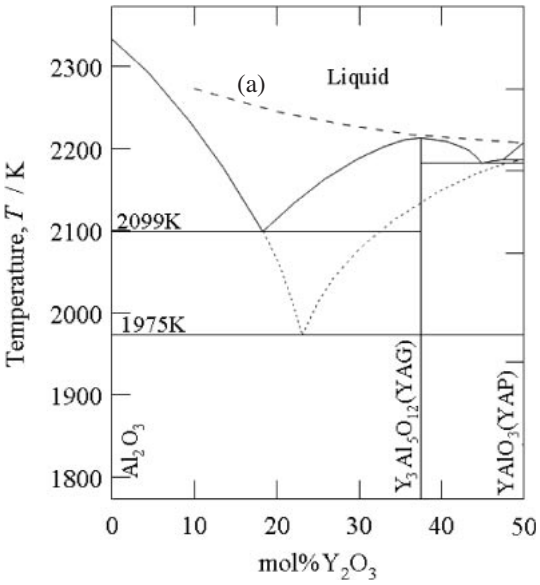

Fig. 1 Phase diagram of the Al_2O_3-Y_2O_3 system. The pecked lines indicate the liquidus of the metastable eutectic system. Melts cooled down from the temperatures above the dashed line (a) follow the metastable path of the solidification.[7)]

in vacuum. The Al_2O_3-YAP metastable eutectic structure was obtained by heating the melt to temperatures above 2300 K before solidification. The solidified specimens were crushed into particles and sieved through 45–63 μm, 32–45 μm and 20 μm sieves in the decreasing order. Two kinds of Al_2O_3-YAP metastable eutectic particles with diameters less than 20 μm and ranging from 20 μm to 45 μm were prepared in this study. The Al_2O_3-YAP particles were inserted into a Mo mold, which has an outer diameter of 10 mm and an inner diameter of 6 mm. The Mo mold was inserted into a carbon die to avoid Joule heat due to direct current in the Mo mold. The direct current through the carbon die was used to heat the specimen. The carbon die was heated to 2023 K at a heating rate of 1.7 K/s under the given pressures (10, 20, 35, and 50 MPa) and cooled after holding at 2073 K for the given holding time (60 s, 10.8 ks, and 21.6 ks). The microstructures of the Al_2O_3-YAG compacts were observed by scanning electron microscopy (SEM). The volume fraction and size of the porosity of the Al_2O_3-YAG compacts were measured using an image analysis program. The flexural strength of the Al_2O_3-YAG compacts was measured using the three-point bending technique over a span of 10 mm and at a crosshead speed of 0.1 mm/min. A rectangular specimen of dimensions $1 \times 2 \times 13\,mm^3$ was used for the flexural strength test. The surfaces of the specimens were polished with #400 emery paper in a direction perpendicular to the flexural axis.

3. Results and Discussions

3.1 Influence of the Al_2O_3-YAP eutectic particle size on the Al_2O_3-YAG microstructure and flexural strength

Figures 2(a) and (b) show the SEM photographs of the Al_2O_3-YAG compacts produced by melting the Al_2O_3-YAP metastable eutectic particles with diameters ranging from 20 μm to 45 μm and less than 20 μm, respectively. The white and black regions in the SEM photographs indicate the YAG phase and the Al_2O_3 phase, respectively. A fine and uniform eutectic structure with a lamellar spacing of 1 μm or less was observed when the Al_2O_3-YAG compact was produced by using the Al_2O_3-YAP metastable eutectic particles with diameters ranging from 20 μm to 45 μm. A eutectic structure in which the lamellar was produced due to coupled growth was recognized in some parts of the compacts. On the other hand, the Al_2O_3-YAG compact produced on using Al_2O_3-YAP particles with diameters less than 20 μm revealed that Al_2O_3 phases with a diameter of 2.5 μm were dispersed in the YAG matrix. The morphology was different from that of the Al_2O_3-YAP eutectic particles with diameters in the range of 20 μm to 45 μm.

It was reported that YAG is likely to nucleate at the Al_2O_3/YAP phase boundary, that volume expansion occurred during phase transformation ($Al_2O_3 + 3YAP = YAG$), and that volume expansion in the solid state suppresses the nucleation of YAG.[6)] Thus, an increase in the three-phase boundary (Al_2O_3/YAP/vacuum) on volume expansion can affect the nucleation and growth of the YAG phase. Consequently, undercooled melt formation can be influenced by the initial Al_2O_3-YAP particle size. In order to clarify the reason why the initial Al_2O_3-YAP particle size resulted in different Al_2O_3-YAG eutectic microstructures, it is important to investigate this phenomenon from the viewpoint of the crystallographic orientation distribution and grain size distribution in the Al_2O_3-YAG eutectic structure.

In the case of the Al_2O_3-YAP metastable eutectic particles with diameters ranging from 20 μm to 45 μm and less than 20 μm, the mean diameters of the porosities were 9 μm and 4 μm, respectively, while the volume fractions of the

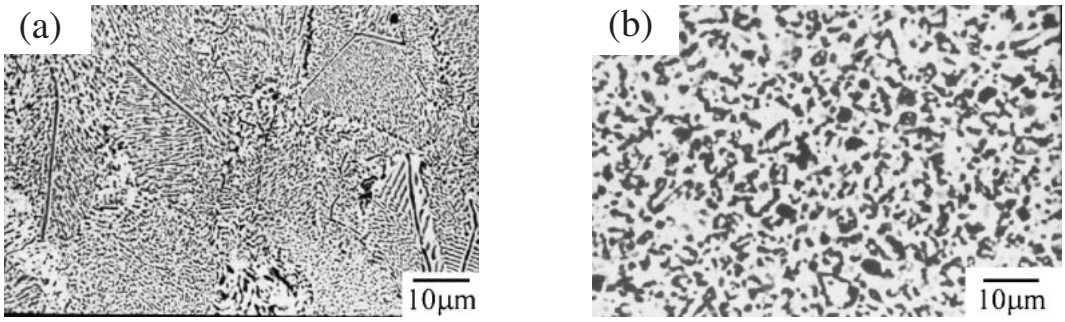

Fig. 2 SEM photographs of the Al_2O_3-YAG compacts (Al_2O_3-23.5 mol%Y_2O_3) produced by the Al_2O_3-YAP particles with diameters ranging (a) from 20 μm to 45 μm and (b) less than 20 μm.

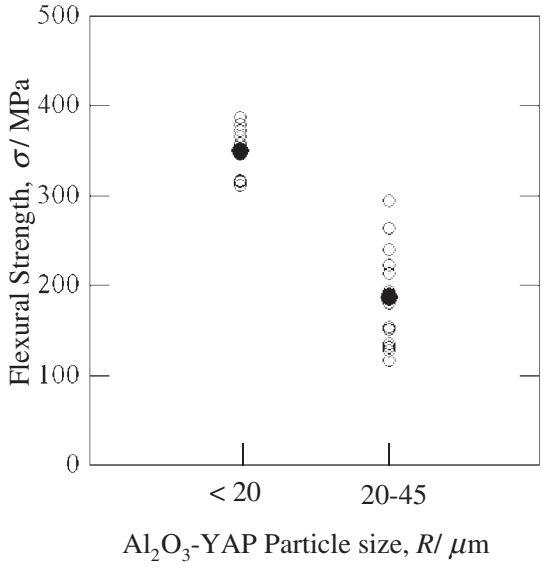

Fig. 3 Flexural strength of the Al_2O_3-YAG compacts (Al_2O_3-23.5 mol%Y_2O_3) produced by the Al_2O_3-YAP particles with diameters ranging from 20 μm to 45 μm and less than 20 μm. The open and closed circles indicate the measured and average values of the flexural strength, respectively.

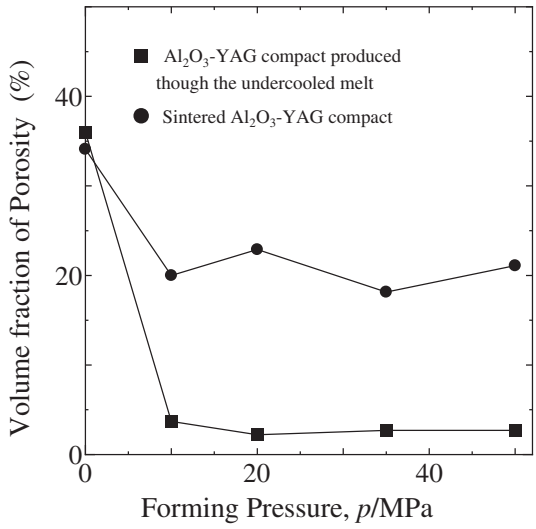

Fig. 4 The forming pressure dependence of volume fraction of the porosity in the Al_2O_3-YAG compact produced using undercooled melt and the sintered Al_2O_3-YAG compact.

porosities were 3% and 2%, respectively. The use of finer Al_2O_3-YAP metastable eutectic particles resulted in a reduction in the size of the porosities.

The flexural strengths of the Al_2O_3-YAG compacts produced from the Al_2O_3-YAP eutectic particles with diameters ranging from 20 μm to 45 μm and less than 20 μm are shown in Fig. 3. The average values of the flexural strength of the compacts produced by using Al_2O_3-YAP eutectic particles with diameters ranging from 20 μm to 45 μm and less than 20 μm were 187 MPa and 339 MPa, respectively. As compared to the Al_2O_3-YAG compact produced from Al_2O_3-YAP particles with diameters ranging from 20 μm to 45 μm, the Al_2O_3-YAG compact produced from the Al_2O_3-YAP eutectic particles with diameters less than 20 μm exhibited a higher flexural strength and lower scatter in the measurement of the flexural strength. This small scatter is due to the smaller porosity size.

The Al_2O_3-YAG compact produced from Al_2O_3-YAP eutectic particles with diameters less than 20 μm exhibits a lower fraction of porosity and higher strength in spite of the coarse eutectic structure. Thus, it is conclude that the Al_2O_3-YAP eutectic particles with diameters less than 20 μm are suitable for producing a dense Al_2O_3-YAG compact with a relatively higher flexural strength. In the following experiments, Al_2O_3-YAP metastable eutectic particles with diameters less than 20 μm size were chosen as the starting materials.

3.2 Influence of forming pressure on the Al_2O_3-YAG microstructure

In order to investigate the influence of the forming pressure on the volume fraction of the porosity, the Al_2O_3-YAG compacts were produced under different forming pressures of 0, 10, 20, 35, and 50 MPa. Figure 4 shows the volume fraction of the porosity in the Al_2O_3-YAG compacts as a function of the forming pressure imposed during shaping. For the purpose of comparison, the volume fraction of the porosity in the sintered Al_2O_3-YAG compacts produced by using Al_2O_3 and YAG particles is also presented. The volume fraction of the porosity of both for the Al_2O_3-YAG compacts formed using the undercooled melt shaping and the sintered Al_2O_3-YAG compact for a forming pressure that exceeded 10 MPa was independent of the forming pressure. The volume fraction in the Al_2O_3-YAG compact produced from Al_2O_3-YAP eutectic particles became less than 4%, while the volume fraction of the porosity of the sintered Al_2O_3-YAG compact was approximately 20%. Undercooled melt shaping achieved a high apparent density even under a relatively low pressure such as 10 MPa. The high volume fraction of the porosity is attributed to the 11% volume expansion in the transformation from the Al_2O_3-YAP system to the Al_2O_3-YAG system.[6)]

3.3 Influence of holding time on the microstructure and flexural strength

The influences of the holding time at a maximum temperature of 2023 K on the microstructure and mechanical property of the Al_2O_3-YAG compacts were examined. SEM photographs of the Al_2O_3-YAG compacts with a holding time of 60 s, 10.8 ks, and 21.6 ks are shown in Figs. 5 (a), (b), and (c), respectively. The values of the mean diameter of the Al_2O_3 grains were 2.5 μm for a holding time of 60 s, 2.7 μm for 10.8 ks, and 4.2 μm for 21.6 ks. The Al_2O_3 phase coarsened with increasing holding time. The volume fractions of the porosity at holding times of 60 s, 10.8 ks, and 21.6 ks were 2%, 2%, and 1%, respectively. The volume fraction of the porosity was independent of the holding time. The connections between the initial particles were completed only for a holding time of 60 s.

The average values of the flexural strength at holding times of 60 s, 10.8 ks, and 21.6 ks were 350, 338, and 268 MPa, respectively. The flexural strength reduced with the increase in the holding time. Mah *et al.* reported that the flexural

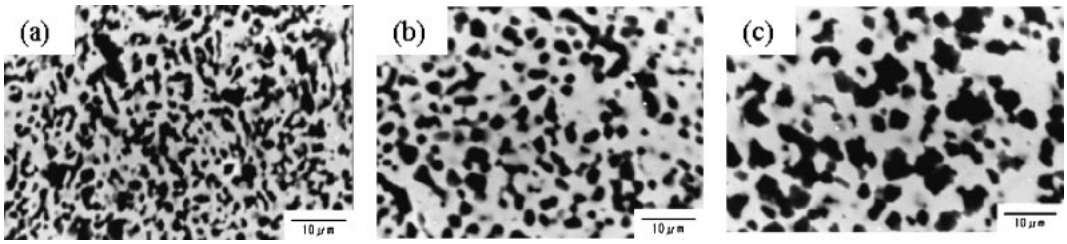

Fig. 5 SEM photographs of the Al_2O_3-YAG compacts kept for holding times of (a) 60 s, (b) 10.8 ks, and (c) 21.6 ks at 2023 K.

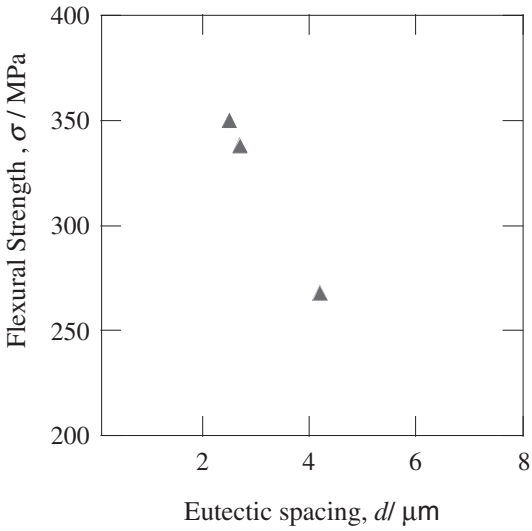

Fig. 6 The flexural strength as a function of eutectic spacing for the Al_2O_3-YAG compacts at holding times of 60 s, 10.8 ks, and 21.6 ks.

strength of the Al_2O_3-YAG system can be determined by the eutectic spacing rather than the grain size.[16,17] Figure 6 shows the flexural strength of the Al_2O_3-YAG compacts at holding times of 60 s, 10.8 ks, and 21.6 ks as a function of the eutectic spacing. The flexural strength has a tendency to depend on the eutectic spacing. Thus, a reduction in the flexural strength is attributable to the increase in the eutectic spacing during holding at the maximum temperature.

Based on these results, the Al_2O_3-YAG compact was immediately formed when undercooled melt was produced at the metastable eutectic temperature. It was not necessary to maintain the compact at the maximum temperature in order to produce a dense compact during undercooled melt shaping.

4. Conclusions

The influences of the initial Al_2O_3-YAP eutectic particle size on the microstructure and mechanical property of Al_2O_3-YAG compact were investigated. Although the Al_2O_3-YAG compact produced from the Al_2O_3-YAP eutectic particles with diameters ranging from 20 μm to 45 μm had a finer eutectic structure as compared to the compact produced from Al_2O_3-YAP eutectic particles with diameters less than 20 μm, utilizing smaller-sized Al_2O_3-YAP eutectic particles provided higher flexural strength. The influence of the forming pressure and holding time at the maximum temperature on the Al_2O_3-YAG microstructure and/or the mechanical property were also examined. The volume fraction of the porosity of the Al_2O_3-YAG compacts was approximately 4% regardless of the forming pressure and holding time. The eutectic spacing increased with the holding time, resulting in a reduction in the flexural strength. In the solidification from the undercooled melt formed by melting the Al_2O_3-YAP eutectic particles with diameters less than 20 μm, a dense Al_2O_3-YAG compact was produced under a low forming pressure of 10 MPa and a short holding time of 60 s.

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REFERENCES

1) T. Mah and T. A. Parthasarathy: Ceram. Eng. Sci. Proc. **11** (1990) 1617–1627.
2) Y. Waku, N. Nakagawa, T. Wakamoto, H. Ohtsubo and K. Shimizu: Nature **389** (1997) 49–52.
3) Y. Waku, N. Nakagawa, T. Wakamoto, H. Ohtsubo, K. Shimizu and Y. Kohtoku: J. Mater. Sci. **33** (1998) 1217–1225.
4) Y. Waku, N. Nakagawa, T. Wakamoto, H. Ohtsubo, K. Shimizu and Y. Kohtoku: J. Mater. Sci. **33** (1998) 4943–4951.
5) T. A. Parthasarathy and T. Mah: J. Am. Ceram. Soc. **76** (1993) 29–32.
6) Randall S. Hay: J. Am. Ceram. Soc. **77** (1994) 1473–1485.
7) J. L. Caslavsky and D. J. Viechinickl: J. Mater. Sci. **15** (1980) 1709–1718.
8) H. Yasuda, I. Ohnaka, Y. Mizutani and Waku: Sci. Tech. Adv. Mater. **2** (2001) 67–71.
9) H. Yasuda, I. Ohnaka, Y. Mizutani and Y. Waku: Proc. M. C. Flemings Symposium TMS. (2001) 171–176.
10) H. Yasuda, T. Mizutani, I. Ohnaka, A. Sugiyama and Y. Waku: Mater. Trans. **42** (2001) 2124–2130.
11) Y. Mizutani, H. Yasuda, I. Ohnaka, A. Sugiyama, S. Takeshima, M. Kirihara and Y. Waku: Mater. Trans. **43** (2002) 2847–2854.
12) Y. Mizutani, H. Yasuda, N. Maeda and Y. Waku: J. Cryst. Growth. **244** (2002) 384–392.
13) H. Yasuda, Y. Mizutani, I. Ohnaka, A. Sugiyama and T. Morikwa: J. Am. Ceram. Soc. **86** (2003) 1818–1820.
14) H. Yasuda, I. Ohnaka, Y. Mizutani, A. Sugiyama, T. Morikawa, S. Takeshima T. Sakimura and Y. Waku: Sci. Tech. Adv. Mater. **5** (2004) 207–217.
15) H. Yasuda, I. Ohnaka, A. Sugiyama, Y. Mizutani, T. Sakimura, A. Kawaguchi and Y. Waku: Mater. Sci. Form. **475–479** (2005) 2709–2712.
16) T. Mah, T. A. Parthasarathy and R. J. Kerans: J. Am. Ceram. Soc. **83** (2000) 2088–2090.
17) T. A. Parthasarathy, T. Mah and L. E. Matson: J. Ceram. Proc. Res. **5** (2004) 380–390.