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Consolidation of Al_2O_3–$Y_3Al_5O_{12}$ (YAG) eutectic powder prepared from induction-melted solid and strength at high temperature

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Abstract

The eutectic composition powder of Al_2O_3–$Y_3Al_5O_{12}$ (YAG) was melted using a Mo crucible by induction heating. The melt was slowly solidified and resulted in a eutectic structure of large twinned crystals. The eutectic solid was ground and sieved into 3–44 μm and 64–124 μm powders. The powder was consolidated to reproduce eutectic composite by the spark plasma system (SPS). Mechanical properties of the consolidated eutectics were measured at room temperature. The high temperature strength was obtained from 1000 to 1400 °C. The consolidated eutectics was not plastically deformed at 1400 °C.

Keywords: Consolidation of eutectic powder; Eutectic composite; High temperature strength; Oxide eutectic composite; Spark plasma sintering; Spark plasma system

1. Introduction

Eutectic composite consists of twinned single crystals.[1–4] The strength of Al_2O_3–ZrO_2 ($8.8Y_2O_3$) eutectic composite fabricated by floating zone method is not decreased near 1600 °C.[2] The single crystals of eutectic composite are responsible for the high temperature strength. MgO–$MgAl_2O_4$, Al_2O_3–$Y_3Al_5O_{12}$ (YAG) and Al_2O_3–$GdAlO_3$ eutectic composites have received attention as heat-resistant materials for turbine blades because of high strength at high temperature.[5–10] These eutectic composites are carefully prepared by unidirectional solidification methods. The crystal size of Al_2O_3–YAG eutectic composites is 20–30 μm in width. The strength of the eutectics consisting of good crystals is not decreased over 1500 °C. A eutectic fiber of Al_2O_3–$Y_3Al_5O_{12}$ (YAG) eutectic composite has been prepared by an edge-defined film-fed growth[11] or by drawing of a laser-heated floating zone.[12] The strength of a fiber composed of crystals of 5–10 μm in width decreases at more than 1200 °C.[13] The process of the fiber production is not regulated with regard to crystal size and defect. Fabrication methods of eutectic composite are not adequate to produce large or near-net shape material. Therefore, solidified eutectic composites are not candidate for high temperature materials such as turbine blade.

Eutectic powders are consolidated by the spark plasma system (SPS)[14–16] and by hot pressing.[17] These eutectic powders are prepared by arc discharge. Arc discharge is a simple and convenient technique, but it gives solids consisting of fine crystals. The structure of a Al_2O_3–YAG eutectic composite grown by unidirectional solidification is the largest crystals and is not changed by heating at 1700 °C for 5 h.[6] Small crystals of the eutectic fiber are obviously grown by heating at 1400 °C.[13] The high temperature stability of eutectic composite is achieved with regard to the largest crystals. The consolidation may be better to be carried out using powders of large crystals.

In this paper, the eutectic composite of Al_2O_3–YAG was melted in a Mo crucible by induction heating and was slowly cooled to enlarge the structure. The cooled solid was crushed and sieved into two powders of 3–44 and 64–124 μm. These particles were consolidated by SPS. Mechanical properties and density of the consolidated eutectic composite were obtained at room temperature, and the strength was measured from 1000 to 1400 °C.

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2. Experimental procedures

Materials were Al_2O_3 (Sumitomo Chemical Co. Ltd, Japan, 99.99%) and Y_2O_3 (Santoku Metal Co., Ltd., Japan, 99.99%) powders. The powders consisting of the eutectic composition (18.4 mol% of Y_2O_3 and 81.6 mol% of Al_2O_3) were blended in ethanol slurry using a polyethylene pot and steel balls coated with polyethylene for 24 h. The blend was dried, molded and fired at 1000 °C for 2 h. The fired compact was melted four times by arc discharge, the top and bottom being reversed.

Pellets melted by arc discharge were loaded into a Mo crucible of 41 mm diameter and 50 mm length. Melting was conducted in a commercial induction heating furnace (Kokusai Electric Co Ltd., Japan, DP-100MP). The Mo crucible was heated up to 2200 °C by induction heating, and slowly solidified at 3–5 °C/min below the melting temperature of Al_2O_3–YAG eutectic composite. Temperatures were measured at the top cover of the Mo crucible.

The solid was ground using a ball mill made of hard metal. The melt was ground and sieved into 3–44 and 64–124 μm powders. The obtained powder was rinsed with a mixture of distilled water, HNO_3 and H_3PO_4 (3:1:1 in volume) to remove the contaminating WC. The consolidation of the pure powder was carried out in a graphite die using the spark plasma system (SPS) (Sumitomo Coal Mining Co. Ltd., Japan, SPS1050). The consolidation temperature was raised from room temperature to 1690–1710 °C for ca. 20 min in a vacuum. The duration of the consolidation temperature was 5–60 min. The pressure for the consolidation was 20 and 40 MPa.

The density of the consolidated powder was determined by Archimedes' principle using water immersion. Elastic modulus of the bulk, 5–10 mm in thickness and 20 mm in diameter, was measured by pulse-echo overlap ultrasonic technique using an ultrasonic detector (Hitachi Kenki Co. Ltd. Japan, ATS-100) and a storage oscilloscope (Iwasaki Tsushinki Co. Ltd. Japan, DS6411). The polished surface was observed with a scanning electron microscope (Hitachi Ltd. Japan, S-3100H) and etched for the observation of fine structures at 1400 °C for 20 min in air. Flexural strength was determined at room temperature using the three point bending test (Shimazu Co. Ltd. Japan, Autograph DSS-500) and from 1000 to 1400 °C using the same type (Autograph AG-20KNG). Sample dimensions were 3 by 4 by 15 mm. The span of lower knife edge was 10 mm. All testing was done at a crosshead speed of 0.5 mm/min. A Vickers hardness tester (Akashi Co. Ltd. Japan, AVK) was used forhardness and toughness measurements of the consolidated eutectics under a load of 98 N. The indentation fracture toughness was calculated from Vickers hardness, Young's modulus, and crack length at indents.[18]

3. Results and discussion

The structures of a solid melted by induction heating is shown in Fig. 1. A white phase is YAG and a black one is alumina. The size of twinned crystals was similar to that of unidirectionally solidified crystals (20–30 μm in width).[6–9] This solidification was a slow cooling method and was not unidirectionally regulated. This cooling method resulted in a colony structure. A colony is formed due to constitutional supercooling. This supercooling is realized when impurity atoms exist or two crystals are present alone. It needs elaborate techniques to fabricate a eutectic composite without the colonies. The growth rate of twinned crystals decreases at the cell boundary, and twinned crystals are coarse on it.[19] The unidirectional solidification is conducted at controlled conditions of cooling rate and direction of crystal growth. In the case of the cooling method, the solidification is not unidirectionally regulated, and its rate is not same in all the melt. Therefore, it is impossible to remove the cell structure. However, it is not important to grow perfect twinned crystals for this study because the eutectic composite is ground into small powders.

The two kinds of particles of 3–44 and 64–124 μm were consolidated by SPS. Some of particles smaller than 44 μm happened to contain one of twinned crystals for the sake of large size of the starting eutectic structure. SPS was developed for sintering of metal and ceramics in the plasma and electric field. Cessation of shrinkage marks the end of sintering and is judged by monitoring a shrinkage meter. The shrinkage monitor is very useful to determine the accurate temperature of sintering and to achieve full densification.[20]

The SEM image of the consolidated eutectics is shown in Figs. 2 and 3. The eutectic structures were not much in the eutectics consolidated from 3–44 μm powders, as shown in Fig. 2. The spaces between the eutectic particles were filled with new crystals. Fig. 3 indicates that fine structures of the eutectics consolidated from 64–124

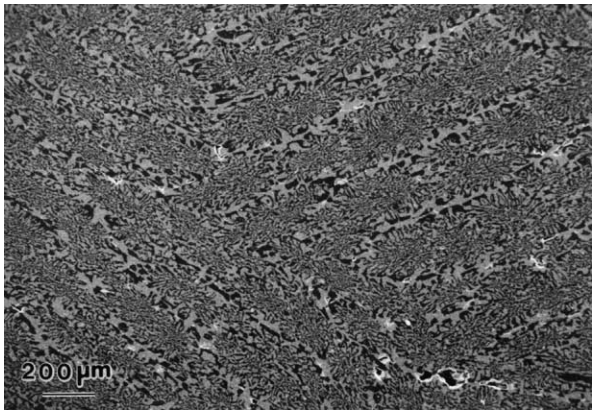

Fig. 1. SEM image of eutectic composite melted by induction heating.

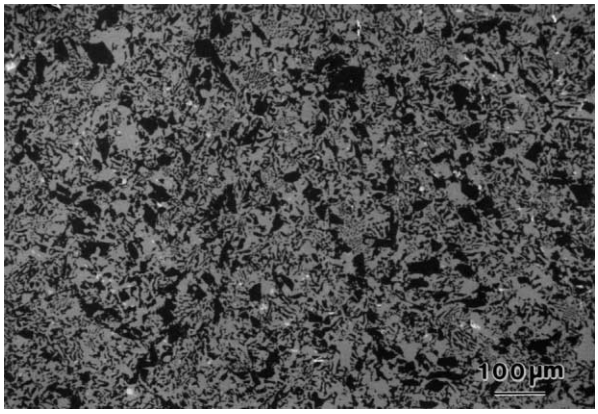

Fig. 2. SEM image of consolidated eutectics from 3–44 μm powder.

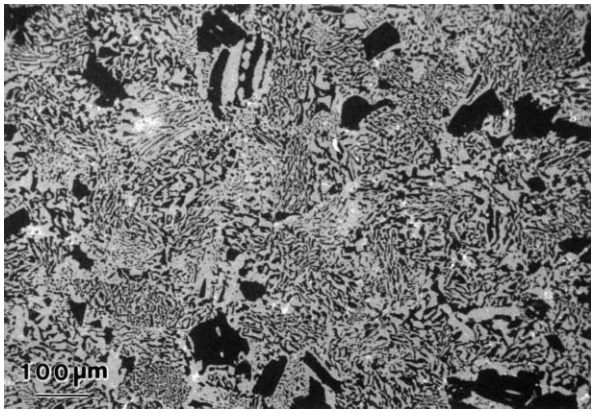

Fig. 3. SEM image of consolidated eutectics from 64–124 μm powder.

μm powders were clarified by etching. The original particle persisted and did not lose its eutectic structure. New crystals were coarse and non-uniform. Especially the eutectics prepared from 64–124 μm powders contained great new crystals. The growth of new crystals was related to the size of the starting particle, and the great crystals appeared with regard to the larger eutectic particles. The thermal-etched surface of the consolidated eutectics is shown in Fig. 4. Grain boundaries were recognized in some of the new crystals. The number of the grain boundary was reduced comparing polycrystal composites. As far as the eutectic powder is starting material, some of twinned crystals are connected with different crystal axes, and the consolidated eutectics has grain boundaries. It is concluded from these

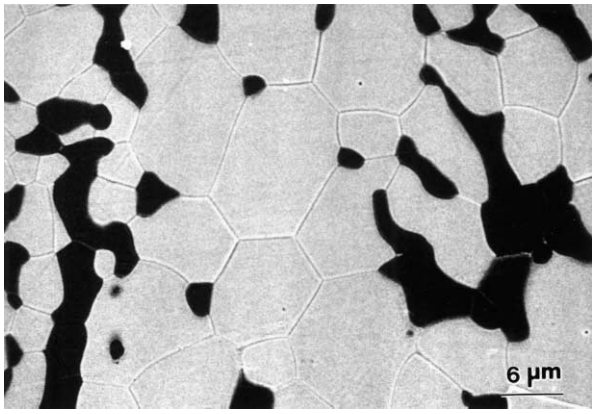

Fig. 4. SEM image of thermal etched surface of consolidated eutectics.

results that the consolidation of eutectic powders can generally accomplish reproduction of eutectic structure.

The properties of the sintered eutectic composites are shown in Table 1, adding those of a polycrystal composite sintered from powders of the eutectic composition. The density more than 99.5% was achieved for the both eutectics prepared from 3–44 and 64–124 μm powders. The consolidation by SPS was useful to produce fully dense materials. The consolidation of large particles seems to be carried out under the effect of plastic deformation. The sintering temperature is near the melting point of the eutectic composites.[16] The deformation of particles accelerates the consolidation and results in the formation of the eutectic composite with high density. Further, the growth of crystals is another important factor in the pulsed electric field. A $CoSb_3$ powder is heated in a graphite die of 20 mm in diameter from 600 to 720 °C at 1 °C/min, and its single crystal of 8 mm in length is obtained by solid-state crystal growth.[20] If the pulsed electric field makes it possible to assist the crystal growth of the twinned crystals, the number of grain boundaries is surely decreased. These two things are advantageous for the reproduction of eutectic composites, diminishing grain boundaries and eliminating pores. Vickers hardness, Young's modulus and Poisson's ratio were almost same for all samples. The indentation toughness of the consolidated eutectics was similar to that of Al_2O_3 and lower than that of the polycrystal composite. The toughness depended on the different structure of the eutectic and polycrystal composites. Grain boundaries

Table 1
Properties of consolidated eutectics and polycrystal composite

Powder (μm)	Relative density (%)	Vickers hardness (GPa)	Fracture toughness ($MPa \cdot m^{1/2}$)	Young's modulus (GPa)	Poisson's ratio	Flexural strength (MPa)
3–44	99.9	16.9±0.3	3.2±0.2	334	0.25	442±8
64–124	99.9	17.1±0.4	2.9±0.1	333	0.25	346±27
Composite[a]	99.5	17.3±0.4	3.6±0.2	334	0.26	621±72

[a] Polycrystal composite.

are responsible for the toughening mechanism of crack deflection, but they are less in the consolidated eutectics than in the polycrystal composite. The strength was obtained by the measurement of the narrow span of 10 mm and tended to be rather large. The space in the furnace to measure the high temperature strength was limited for the span of 10 mm. The strength measured for 30 mm span was about 20% lower than that for 10 mm span at room temperature. The strength was related to the size of the grain structure. The fines grain of the polycrystal composite resulted in the large strength. The strength of the eutectics prepared from 64–124 μm powders was the lowest in all samples. These results indicate that large grains include large defects inside the consolidated eutectics. However, the strength of 346 MPa might not be achieved for a polycrystal composite prepared from the big particle such as 124 μm. It is not clarified whether consolidating eutectic particle results in decreasing crack length or the consolidation by SPS can lower crack size.

The strength at high temperatures was measured from 1000 to 1400 °C in a vacuum, as shown in Fig. 5. The stress-displacement curve of each measurement is cited in Fig. 6. The polycrystal composite started to lose its strength at 1000 °C and was plastically deformed at 1400 °C. The strength of the consolidated eutectics was decreased from 1200 °C. The strength at 1400 °C was less than half that at room temperature. The eutectic composite prepared by the unidirectional solidification is not deteriorated near 1400 °C.[5–10] There is a reason why the consolidated eutectic loses its strength at 1400 °C. The new crystals are not a complete eutectic structure, and the part of them is rather similar to the structure of a polycrystal composite. The ratio of new crystals is larger for the consolidated eutectics prepared from 3–44 mm powders than for that prepared from 64–124 μm powders, as shown in Figs. 2 and 3. The high temperature strength is greatly decreased for the consolidated eutectics fabricated from the smaller powder. The eutectic structure is responsible for the strength at high temperature. It is recommended to use particles containing eutectic structure to retain strength at high temperature.

4. Summary

A eutectic composite of Al_2O_3–$Y_3Al_5O_{12}$ (YAG) was prepared by induction heating. The eutectic solid was ground and sieved into 3–44 and 64–124 μm powders. These powders were consolidated by the spark plasma system (SPS). New crystals were formed during the consolidation of the eutectic powder. The part of the new crystals contained grain boundaries, but the consolidation of the eutectic powder made it possible to roughly reproduce eutectic structure. The strength was measured from room temperature to 1400 °C. The strength of the consolidated eutectics was 424 and 328 MPa for the eutectic powders of 3–44 and 64–124 μm at room temperature, respectively. Plastic deformation was not observed on the stress-displacement curve of the

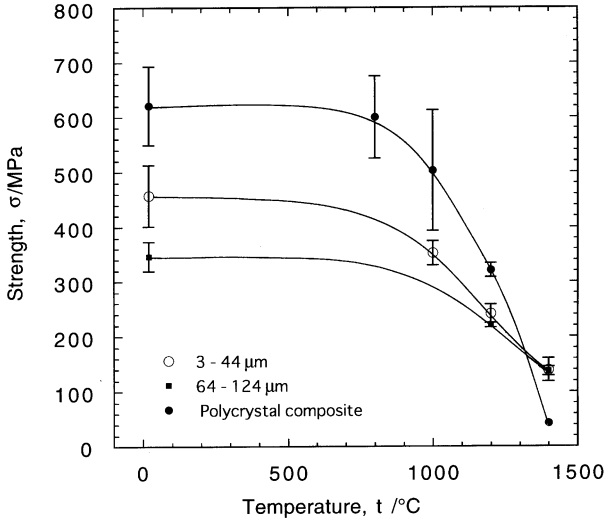

Fig. 5. Strength of consolidated eutectics and polycrystal composite at room and high temperatures.

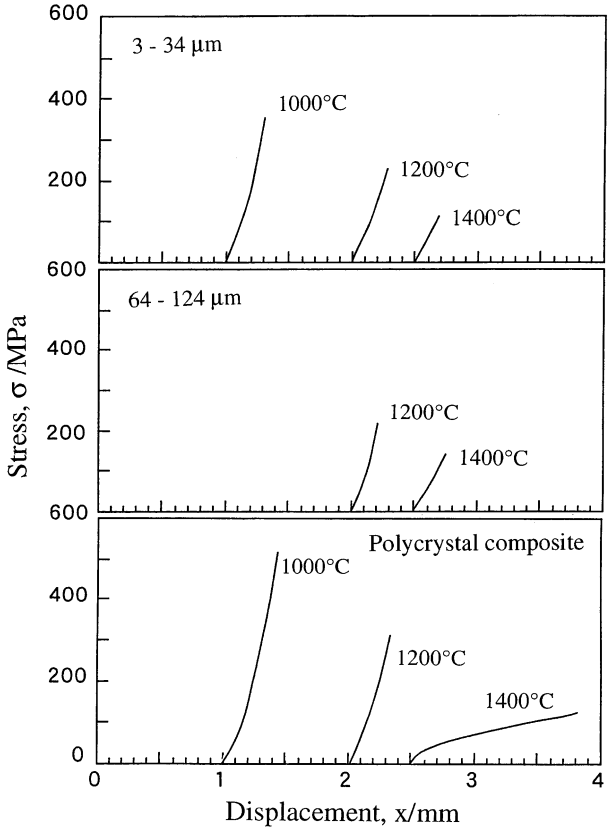

Fig. 6. Stress-displacement curve of consolidated eutectics and polycrystal composite.

consolidated eutectics at 1400 °C, but the high temperature strength was decreased less than half that at room temperature.

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