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Dislocation mechanism of deformation and strength of Al_2O_3–YAG single crystal composites at high temperatures above 1500°C

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Abstract

A new unidirectionally solidified eutectic Al_2O_3–YAG composite has recently been fabricated by accurately controlling the unidirectional solidification. The eutectic composite has a new microstructure, in which single crystal Al_2O_3 and single crystal YAG are three-dimensionally and continuously connected and finely entangled without grain boundaries. The dislocation structure is observed in both single crystal Al_2O_3 and YAG in the plastically deformed specimens in the tensile and compressive tests at high temperatures for the Al_2O_3–YAG single crystal composite, showing that the plastic deformation occurred by dislocation motion. The Al_2O_3–YAG single crystal composite fabricated has the following properties: (1) the flexural strength at room temperature can be maintained up to just below melting point (about 1830°C), (2) the compressive flow stress at 1600°C and a strain rate of 10^{-4}/s is about 13 times higher than that of sintered composites of the same composition.

Keywords: Al_2O_3–YAG; Composites; Dislocations; Microstructure-final; Strength

1. Introduction

Recently all industrial fields are expected to improve the efficiency of energy consumption. To make sound, sustainable economic growth with compatibly efficient use of energy, it is necessary for the energy conversion efficiency of power-generation systems and power systems to be rapidly improved. For example, to improve the combustion efficiency of gas turbines, operating temperatures must be increased and to achieve this, the development of ultra-high-temperature resistant structural materials is indispensable. Currently Ni-base superalloys are the main thrust in this field, but these have melting points of less than 1400°C, and their strength deteriorates sharply in the region of 1000°C. For this reason, in recent years in order to overcome the heat resistance limitations of metals, the development of turbine technology using advanced materials, centered on ceramic composites, has been vigorously pursued.

The recently-developed Al_2O_3–YAG eutectic material[1–3] is a new ceramic composite made by melting and unidirectional solidification of raw material oxides using a eutectic reaction to precisely control the crystal growth. In this paper, the microstructural and high temperature characteristics such as the temperature dependence of flexural strength, the tensile and compressive plastic deformation behavior of the unidirectionally solidified Al_2O_3–YAG eutectic composite are reviewed and discussed.

2. Manufacturing process

Most conventional ceramics and ceramic composites are produced by using the powder sintering method. For this reason, they contain many impurities, and there are many cases of amorphous phases forming at grain boundaries. In many cases, these amorphous phases and grain boundaries effectively act to increase fracture toughness and strength at room temperature, but there are microstructural factors thought to have a deleterious effect on high-temperature strength and creep characteristics. The Al_2O_3–YAG eutectic composite eliminates these microstructural factors, by precisely controlling its stable high-temperature microstructure.[1–3] In the case of the Al_2O_3–YAG eutectic composite manufacturing process, raw material powders are pre-melted to obtain an ingot, and then the ingot is placed in a

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molybdenum crucible. The melting experiment was performed in the Mo crucible heated by high-frequency induction heating, and then, after holding 30 min at 1950°C, unidirectional solidification was carried out by lowering the Mo crucible at a speed of 5 mm/h.[1–3] As outlined, the Al_2O_3/YAG eutectic composite manufacturing process is completely different from the conventional powder sintering method, and the basic manufacturing principle resembles the production method of Ni-based single-crystal cast superalloys.

3. Microstructure of Al_2O_3–YAG eutectic composite

The microstructures of the upper, middle and lower planes perpendicular to the solidification direction of the Al_2O_3–YAG eutectic composite, and those of the hot-pressed plane of the sintered composite with the same composition, consisted of Al_2O_3 and YAG phases; these were determined from X-ray diffraction patterns.[1–3] Fig. 1(a) and (b) show SEM images of the cross-section perpendicular to the solidification direction of the eutectic composite and parallel to the hot-pressed plane of the sintered composite. The light area in the SEM micrograph is the YAG phase, and the dark area is the Al_2O_3 phase (identified by EPMA analysis). Homogeneous microstructures with no pores or colonies are observed in the eutectic composite (Fig. 1a). In the X-ray diffraction pattern for the Al_2O_3–YAG eutectic composite, diffraction peaks from the (743) plane of the YAG phase in the plane perpendicular to the solidification direction and from the (110) plane of the Al_2O_3 phase in the plane inclined by 76° from the solidification direction are observed. Consequently, it can be concluded that this composite consists of $<110>$ single-crystal Al_2O_3 with a hexagonal structure and $<743>$ single-crystal YAG with a garnet structure. In contrast, for the sintered composite, diffraction peaks from various planes are observed, characteristic of a polycrystalline ceramic composite with random crystal orientations. Fig. 2 shows an SEM micrograph which illustrates the three-dimensional configuration of the single-crystal YAG in the Al_2O_3–YAG eutectic composite from which Al_2O_3 phases had been removed by heating in graphite powder at 1650°C for 2 h. The single-crystal YAG is a three-dimensionally connected, extremely complex, having a hieroglyphic-like configuration. We therefore conclude that the present Al_2O_3–YAG eutectic composite has a microstructure consisting of three-dimensionally continuous and complexly entangled single-crystal Al_2O_3 and single-crystal YAG.

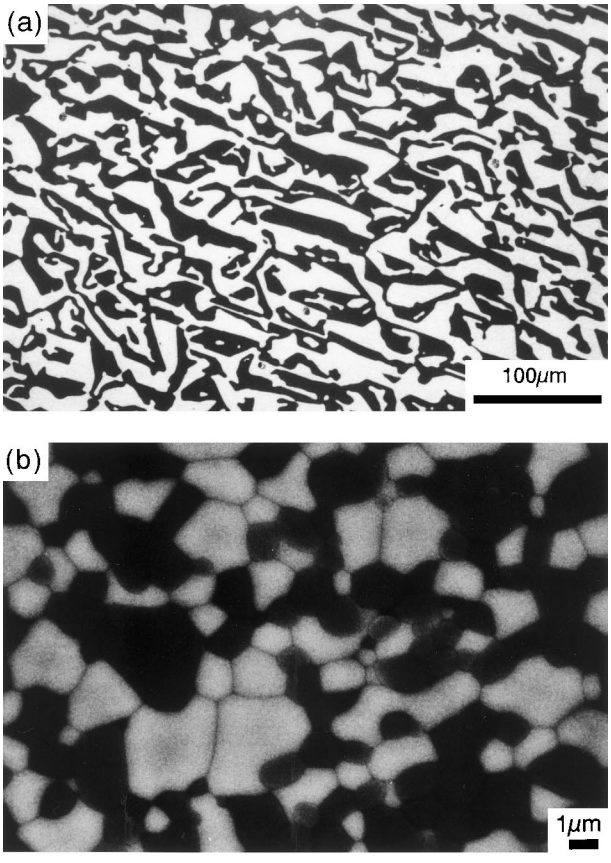

Fig. 1. (a) SEM showing the microstructure of a cross-section perpendicular to the solidification direction of the Al_2O_3–YAG eutectic composite; (b) SEM image showing the microstructure of a cross-section parallel to the hot-pressed plane of the sintered Al_2O_3–YAG composite.

4. Temperature dependence of flexural strength

Fig. 3 shows the temperature dependence of the flexural strength of an Al_2O_3–YAG single crystal composite from room temperature to 1800°C in comparison with that of a sintered composite of the same composition.[3] The Al_2O_3–YAG single crystal composite maintains its

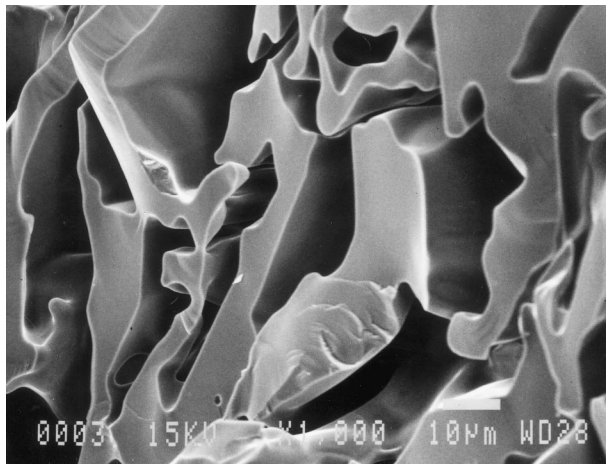

Fig. 2. SEM micrograph showing the three-dimensional configuration of single-crystal YAG in the Al_2O_3–YAG eutectic composite.

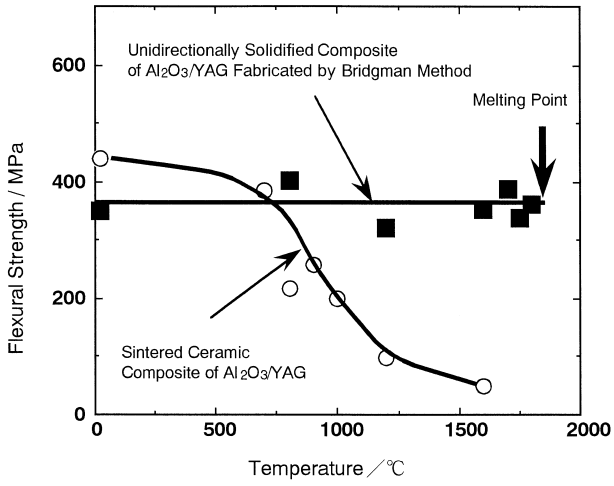

Fig. 3. Temperature dependence of flexural strength of Al_2O_3–YAG single crystal composites compared with sintered composites.

room temperature strength up to 1800°C (just below its melting point of about 1830°C), with a flexural strength in the range of 350–400 MPa. The sintered composite, on the other hand, has the same or higher flexural strength at room temperature, but its strength falls precipitously above 800°C.

Sintered composites show intergranular fracture at room temperature and at 1400°C and evidence for grain growth is clear.[3] On the other hand, the Al_2O_3–YAG single crystal composites show no grain growth up to the very high temperature of 1700°C, and the fracture is transgranular.[3] Moreover, when the test temperature reaches 1800°C, the fracture at the interface between the Al_2O_3 and YAG phases and mixed fracture of intergranular and transgranular is observed.[3]

Generally, if an interface or a grain boundary contain an amorphous phase, high-temperature strength is reduced.[4,5] Fig. 4 shows HRTEM images of the grain boundaries between the Al_2O_3 and YAG phases of a sintered composite and the interface between the Al_2O_3 and YAG phases of an Al_2O_3–YAG single crystal composite. As we can see from Fig. 4(a), the sintered composite interface contains an amorphous phase. However, as it is evident from Fig. 4(b), the interface between the Al_2O_3 and YAG phases in the Al_2O_3–YAG single crystal composite contains no amorphous phase.

From the above, it may be concluded that the superior high temperature strength was obtained by the following means: good crystal orientation of matrix, consisting of $<110>$ single crystal Al_2O_3 and $<743>$ single crystal YAG; no amorphous phases formed at interface between Al_2O_3 phases and YAG phases, which can easily cause plastic deformation; and the effect of the eutectic composite consisting of single crystal Al_2O_3 and YAG, which are stable at very high temperatures.

5. Tensile deformation

Fig. 5 shows the nominal tensile stress-elongation curve obtained from tensile tests of an Al_2O_3–YAG single crystal composite from room temperature to 1750°C.[3] Above 1650°C a yield phenomenon occurs and the composites fracture after around 10–17% plastic deformation. The yield stress is about 200 MPa at 1650°C. Several cracks appeared in the microstructure at both the 1650 and 1750°C temperature levels. Nearly all of the cracks were in the YAG phase, with almost none observed in the Al_2O_3 phase. A SEM observation

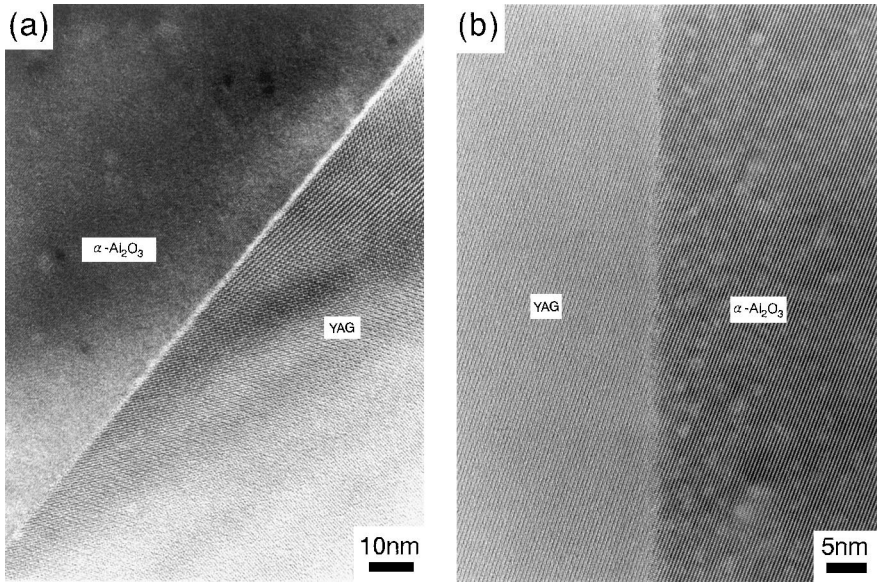

Fig. 4. HRTEM images of the grain boundaries between the Al_2O_3 and YAG phases in a sintered composite and (b) the interface between Al_2O_3 and YAG phases of Al_2O_3–YAG single crystal composite.

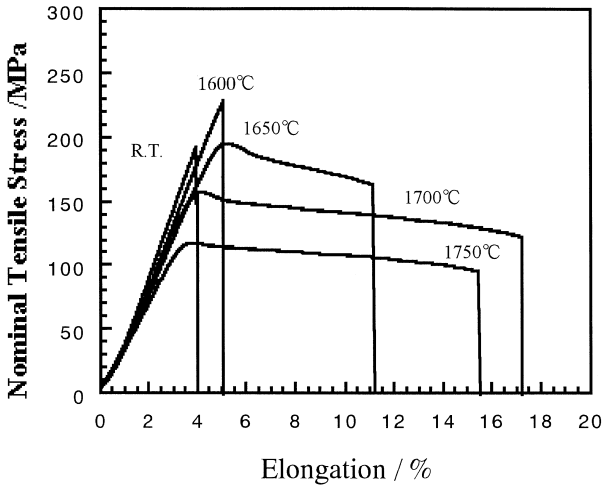

Fig. 5. Nominal tensile stress-elongation curves of an Al_2O_3–YAG single crystal composite from room temperature to 1750°C.

of the fracture surface at tensile testing reveals a constricted area in which a ductile fracture can be observed in the Al_2O_3 phase. In a part of the image, dimple-shaped fracture surface can also be observed. Also, the type of fracture is mixed; intergranular and transgranular fracture are both present.[3]

Fig. 6 shows bright field TEM images of dislocation structures observed in the plastically deformed specimen in the tensile test at 1700°C for the Al_2O_3–YAG single crystal composite. Though the dislocation structures are to be observed in both single crystal Al_2O_3 and single crystal YAG, showing that the plastic deformation occurred by dislocation motion, dislocation densities and dislocation structures in both phases are largely different. Namely, many linear dislocations are observed in single crystal Al_2O_3. Meanwhile, low dislocation density is observed in single crystal YAG.

6. Compressive deformation

Fig. 7 shows the relationship between compressive flow stress and the strain rate in an Al_2O_3–YAG single crystal composite and a sintered composite at test temperatures of 1500, 1600, and 1700°C.[6] While the Al_2O_3–YAG single crystal composite and the sintered composite shared the same chemical composition and constitutional phases, their compressive deformation was markedly different. That is, at the same strain rate of 10^{-4}/s and test temperature of 1600°C, the sintered composite showed a flow stress of 33 MPa, while the Al_2O_3–YAG single crystal composite's flow stress was approximately 13 times higher at 433 MPa. Moreover, as can be seen from the diagram, the Al_2O_3–YAG single crystal composite has creep characteristics that surpass those of *a*-axis sapphire fibers[7] and, as a bulk material, displays excellent creep resistance.

Fig. 8 shows the bright field TEM images of dislocation structure observed in the specimens plastically

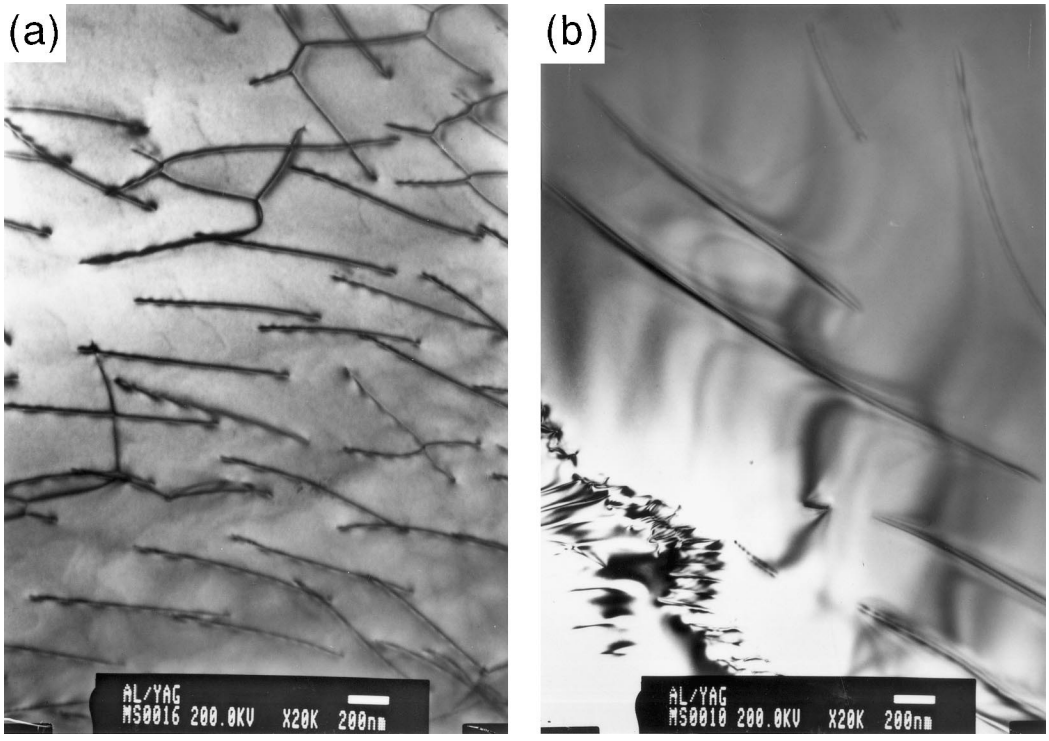

Fig. 6. TEM images showing the dislocation structures of (a) Al_2O_3 phases and (b) YAG phases of the plastically deformed specimens after the tensile test at 1700°C of the Al_2O_3–YAG single crystal composite.

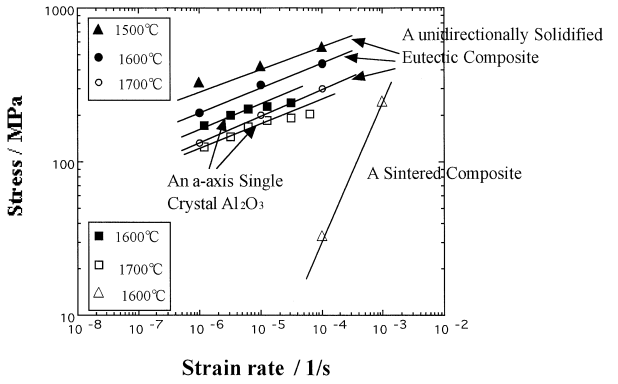

Fig. 7. Relationship between compressive flow stress and strain rate for an Al_2O_3–YAG single crystal composite, a sintered composite and an *a*-axis Al_2O_3 single crystal.

deformed around 14% in the compressive test at an initial strain rate of 10^{-5}/s and test temperature of 1600°C for an Al_2O_3–YAG single crystal composite and a sintered composite. Dislocation structure is observed in both Al_2O_3 phase and YAG phase for the Al_2O_3–YAG single crystal composite, showing that the plastic deformation occurred by dislocation motion. While dislocation was not observed in both Al_2O_3 phase and YAG phase for the sintered composite. The dislocation structures observed in the Al_2O_3–YAG single crystal composite also indicate that the plastic deformation mechanism of the present eutectic composite is essentially different from that of the sintered composite similar to the micrograin superplasticity of ceramics[8] due to a grain-boundary sliding or a liquid phase present at grain boundary at a high temperature.

The steady state creep rate $\dot{\varepsilon}$, can be usually shown by the following equation:

$$\dot{\varepsilon} = A\sigma^n \exp(-Q/RT) \quad (1)$$

Here, A and n are dimensionless coefficients, σ is the creep stress, Q is the activation energy for the creep, T is the absolute temperature, while R is the gas constant.[9] In Fig. 7, the value of n is around 1–2 for sintered composites, and 5–6 for Al_2O_3–YAG single crystal composites. In sintered composites, it can be assumed that the creep deformation mechanism follows the Nabarro–Herring or Coble creep models, while in Al_2O_3–YAG single crystal composites, the creep deformation mechanism can be assumed to follow the dislocation creep models corresponding to the dislocation structure in Fig. 8.[6] The activation energy Q is estimated to be about 700 kJ/mol from an Arrhenius plot,[10] which is not so different from the values estimated from the high temperature creep in Al_2O_3 single crystal (compression axis is [110]) and YAG single crystal (compression axis is [110]).[7,11,12] It is also reported that the activation energy for oxygen diffusion in Al_2O_3 is about 665 kJ/mol,[12] which is not so far from the activation energy of Al_2O_3 single crystal for plastic flow even though that of Al^+ diffusion is about 476 kJ/mol.[13] This fact means that the deformation mechanism of the Al_2O_3 single crystal is the diffusion controlled dislocation creep. On the other hand, the activation energy for oxygen diffusion in YAG is about 310 kJ/mol,[14,15] which differs significantly from the activation energy of YAG single crystal for plastic flow. However, dislocation is always observed in both Al_2O_3 phase and YAG phase of compressively deformed specimens at 1500°C–1700°C and at strain rate of 10^{-4}/s–10^{-6}/s. Therefore, the compressive deformation mechanism of the Al_2O_3–YAG single crystal composite must follow the dislocation creep models.

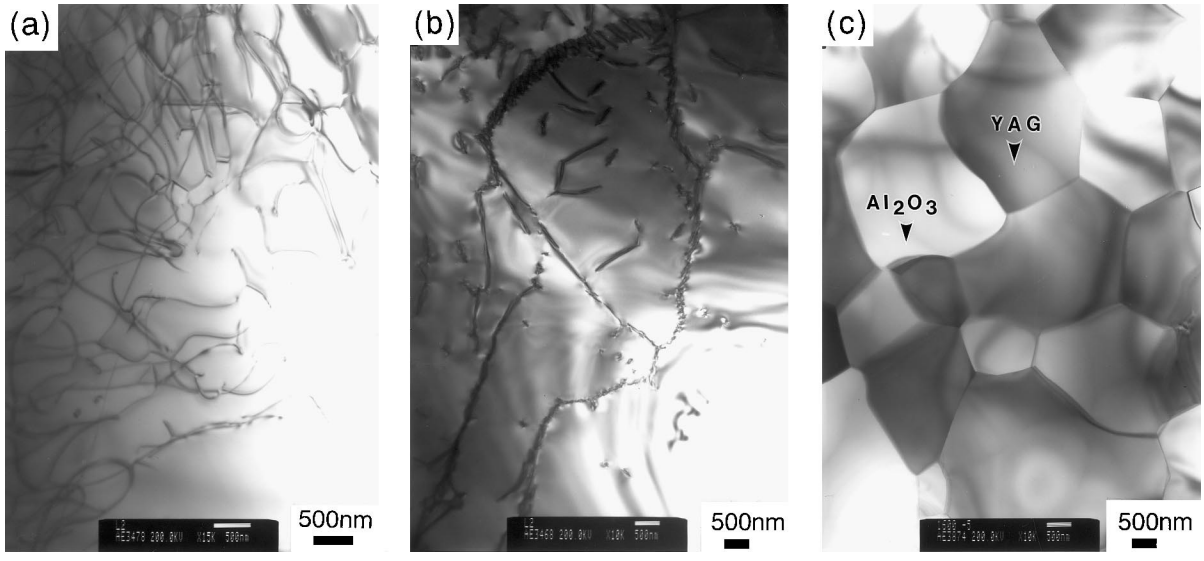

Fig. 8. TEM images showing the dislocation structure of (a) Al_2O_3 phases and (b) YAG phases in the Al_2O_3–YAG single crystal composite , and (c) the microstructure of Al_2O_3 and YAG phases in the sintered composites, of compressively crept specimens at 1600°C and strain rate of 10^{-5}/s.

7. Conclusions

Employing unidirectional solidification, an Al_2O_3–YAG eutectic composite, 40 mm diameter by 70 mm length, was manufactured successfully. The microstrutural and high-temperature characteristics such as the three-dimensionally configuration of constitutional phases, the temperature dependence of flexural strength, the tensile and compressive plastic deformation behavior of the Al_2O_3–YAG eutectic composite are investigated.

This composite has a microstructure in which continuous networks of single-crystal Al_2O_3 and single-crystal YAG interpenetrate without grain boundaries. The Al_2O_3–YAG single crystal composite has superior high-temperature strength characteristics with flexural strength showing no temperature dependence in the range from room temperature up to 1800°C. In tensile test at above 1650°C, the composite showed marked plastic deformation occurring by dislocation motion.

The compressive flow stress of the Al_2O_3–YAG single crystal composite was about 13 times higher than that of a sintered composite with the same chemical composition. The stress exponent was about 1–2 for the sintered ceramics, about 5–6 for the Al_2O_3–YAG single crystal composite. The activation energy for the creep of the Al_2O_3–YAG single crystal composite was around 700 kJ/mol.

Dislocation structures were observed both in Al_2O_3 and YAG phases showing that the plastic deformation occurred by dislocation motion, in the compressively deformed specimens for Al_2O_3–YAG single crystal composite. While dislocation is not observed in both Al_2O_3 phase and YAG phase for the sintered composite. Therefore, the compressive deformation mechanism of the Al_2O_3–YAG single crystal composite must be the dislocation creep.

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