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High Temperature Mechanical Behavior of $MgAl_2O_4$-YAG Eutectic Ceramic *In Situ* Composites by Float Zone Method

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Abstract: The directionally solidified eutectic $MgAl_2O_4$-$Y_3Al_5O_{12}$ crystal was prepared at a pressure of 0.4 MPa of ambient nitrogen gas by the high frequency induction heated floating zone furnace. In order to determine the high temperature characteristics, directionally solidified $MgAl_2O_4$-$Y_3Al_5O_{12}$ eutectic phase has been analyzed with creep test, tensile strength, young modulus and fracture toughness at the various temperatures and the microstructural variations have been studied according to the analysis results. It has been seen that directionally solidified with zone melting $MgAl_2O_4$-YAG eutectic ceramic which has given the value of 168 MPa below 10^{-6}/s strain rate at 1,700 °C temperature has revealed minimum stress.

Keywords: directionally solidified, eutectic, high temperature, mechanical behavior, oxide ceramic materials

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Introduction

Directionally solidified eutectic ceramic materials are used to obtain a material that is protecting its mechanical properties at high temperatures. These materials obtained in studies recently are usually composed of the systems of $Al_2O_3/Y_3Al_5O_{12}$, $Al_2O_3/GdAlO_3$, $Al_2O_3/Er_3Al_3O_{12}$ and Al_2O_3/EAG/ZrO_2 [1, 2]. These materials are candidates for ultra-high temperature materials that will be used in power generation and aerospace industries in the future. Especially, these materials have significant advantages over conventional structural ceramic materials at high temperatures. Directionally solidified eutectic ceramics materials do not contain grain boundary or colonies causing a decrease at mechanical properties at high temperatures. Increasing thermal efficiency of gas turbine engine means to working at high temperatures. But carbides, nitrides and Ni-based superalloys can not maintain their stabilities especially at temperatures higher than 1,500 °C [3]. Currently, one of the most common methods for directionally solidification of oxide ceramics is float zone method [4–6]. The advantage of eutectic single crystal structures with respect to single phase crystals like sapphire is to form an interface that is both though and strength as a result of interaction of both phases. There are some advantages of this solidification made of melt between liquid and solid like control of impurities during growth, decrease of contamination coming from crucible, less energy, providing a uniform doping with zone refining and control of surface tension. It has been reported [7–9] that directionally solidified $MgAl_2O_4$-$Y_3Al_5O_{12}$ ceramics exhibit mechanical behaviors similar to oxide materials used in previous studies [10–33]. But mechanical behaviors of directionally solidified $MgAl_2O_4$-$Y_3Al_5O_{12}$ ceramics at high temperatures have not been reviewed in detail. In this study, it has been examined whether these ceramics meet expected properties at high temperatures.

Experimental

$MgAl_2O_4$-YAG ($Y_3Al_5O_{12}$) samples were prepared by mixing of the Al_2O_3 (99.99 %, Aldrich), MgO (99.99 %, Aldrich) and Y_2O_3 (99.999 %, Alfa) starting powders, and contained 55/25 mass % Al_2O_3/MgO [33]. As it has been shown in Figure 1, directionally solidified eutectic $MgAl_2O_4$ (MAS, magnesium aluminate spinel)-YAG of 10 mm in diameter and 200 mm in length was grown 1 mm/min from the precursor rod by the float zone method. Compression creep tests were performed in an argon atmosphere at a constant cross head speed, using a servohydraulic testing machine. The compression tests were conducted at 1,500 °C–1,700 °C. The rectangular samples with the dimension of 5 mm (width) × 2 mm (height) × 48 (length) mm were cut perpendicular to the direction of solidification using a diamond saw. Tensile strength were

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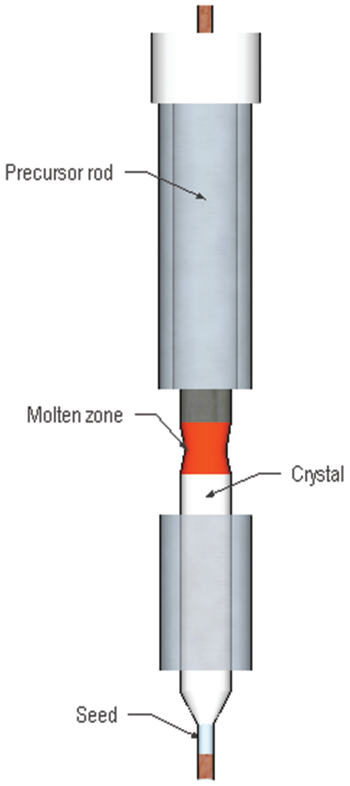

Figure 1: Schematic drawing of the float zone type unidirectional solidification apparatus.

examined at a strain rate of 1×10^{-4} s^{-1} from room temperature to 1,500 °C. The facilities used in the tests were the same machine for compression creep test. The dimensions of the tensile test sample were shown in Figure 2. The elastic constant of MAS-YAG samples were evaluated at various temperatures using the Impulse Excitation Technique by an IMCE machine (RFDA 23, HT 1600). The elastic modulus measurement has been performed according to ASTM E1876-99 [34]. The fracture toughness was evaluated by a single-edge notched beam (SENB) method with a span of 16 mm and a cross head speed 0.05 mm/min. The eutectic composite was cut into bar (3 mm × 4 mm × 35 mm) specimens for fracture toughness testing. The specimen was notched with a diamond cut. The MAS-YAG crystal was located outside the heating chamber [35,36]. The microstructure of the eutectic ceramics were obtained by scanning electron microscope (SEM). The elemental analysis was measured by energy dispersive X-ray spectroscopy (SEM-EDS). Dislocation structures were observed by transmission electron microscopy (TEM).

Results and discussion

Creep behavior

The compressive stress (σ)– strain ($\dot{\varepsilon}$) curves of the MAS-YAG samples are given in Figure 3 as a function of temperature. The creep data were analyzed using power-law relationship:

$$\dot{\varepsilon} = A\sigma^n \exp\left(\frac{Q}{RT}\right) \tag{1}$$

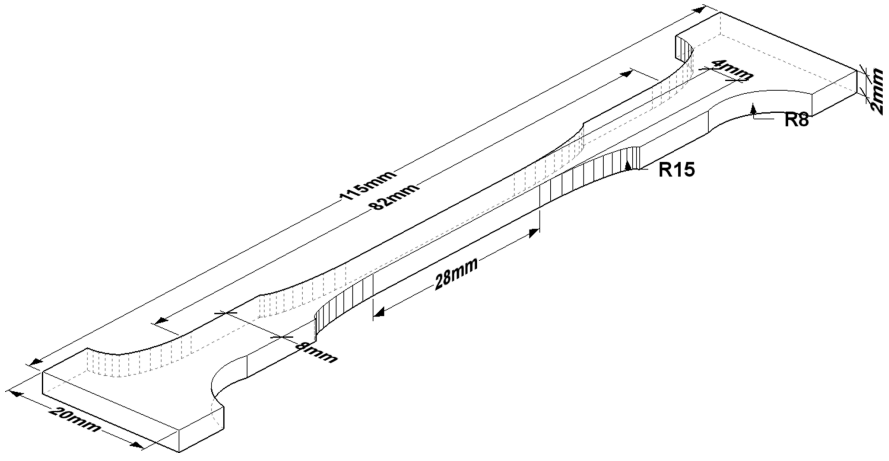

Figure 2: Dimensions of the tensile test specimen.

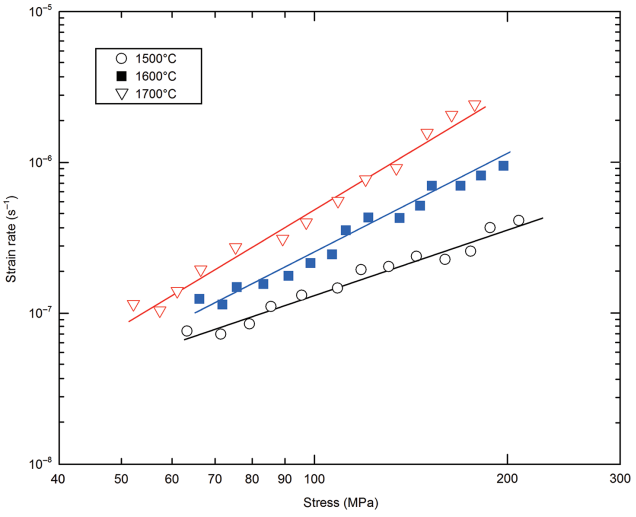

Figure 3: The compressive strain rate-stress behavior as a function of temperature for $MgAl_2O_4$-$Y_3Al_5O_{12}$ crystal.

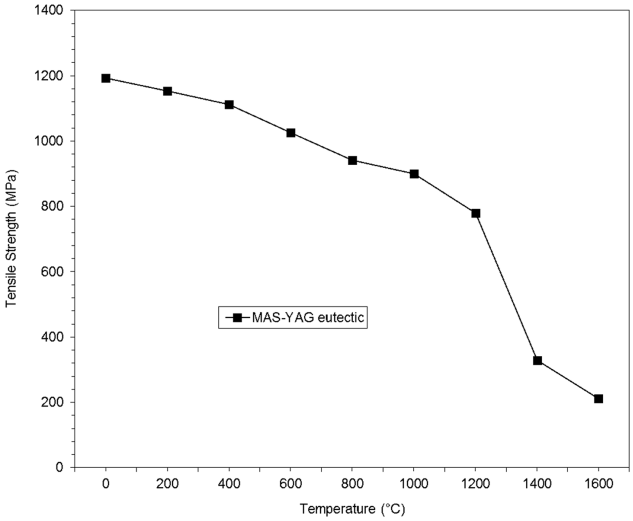

Figure 4: Temperature-dependent tensile strength of $MgAl_2O_4$-$Y_3Al_5O_{12}$.

Here, A is a constant, k the Boltzmann's constant, T the absolute temperature, σ the creep stress, n the stress exponent, Q the activation energy of an Arrhenius temperature dependence and R the gas constant [37]. The stress dependence of strain rates at 1,500 °C, 1,600 °C and 1,700 °C is almost similar for the $MgAl_2O_4$-$Y_3Al_5O_{12}$ crystal. The creep rate of MAS-YAG eutectic at 1,700 °C exceeds that of MAS-YAG eutectics at 1,600 and 1,500 °C. Under the same stress, the strain rate of MAS-YAG eutectic at 1,700 °C is slightly higher than that of 1,600 °C and 1,500 °C. The results indicate that the creep strength of MAS/YAG ceramics was increased insignificantly by 2.4 %, and 5.2 % when the temperatures were increased from 1,500 °C to 1,600 °C, and from 1,600 °C to 1,700 °C, respectively. Elevated temperatures cause no significant change in the creep resistance. For traditional sintered ceramics, increase of temperature decreases creep resistance. But for directionally solidified MAS/YAG ceramics, specific structure of interface between two phases prevents crack propagation. Because this region enables dislocation motion as well as restricts crack growth.

Tensile properties

The tensile strength-temperature curve is shown in Figure 4. It can be seen that tensile strength decreases gradually with increasing in temperature. MAS-YAG structure maintains its ambient temperature strength up to 1,200 °C. The tensile strength dropped rapidly above 1,200 °C. The strength of directionally solidified MAS-YAG (DSMY) sample is higher than the Al_2O_3-YAG (≈780 MPa) structure [38] for 1,200 °C. The strength of DSMY at 1,400 °C was significantly lower than at 1,200 °C. It is obvious that temperature dependent tensile strength properties are effective on ductility of MAS-YAG crystal. At 1,200 °C, MAS-YAG crystal having higher tensile strength than directionally solidified Al_2O_3-YAG composite shows that MAS-YAG crystal eliminates the stress exposed under high temperature but at the same time it preserves its strength.

Temperature dependence of Young's modulus

The Young's moduli of MAS-YAG sample of 10 mm (width) × 5 mm (height) × 48 mm (length) were measured by resonant frequency damping analyzer (RFDA). The Young's modulus of the MAS-YAG sample calculated as [39,40].

$$E = 0.9465 \frac{m f_f^2}{b} \frac{l^3}{d^3} T_1 \quad (2)$$

where m is the mass of sample, f_f is the resonant frequency, b, d and l are the width, thickness and length of the MAS-YAG structure respectively, and T_1 is a correction factor. Figure 5 shows the temperature dependence of elastic modulus directionally solidified MAS-YAG crystal. The elastic modulus of MAS-YAG decreased as temperature increased. At 1,200 °C the Young's moduli were

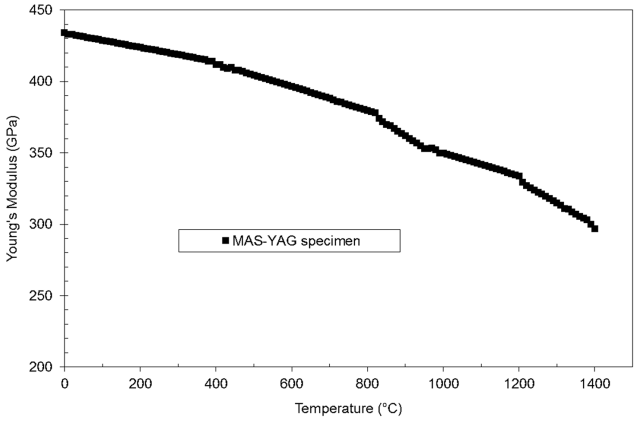

Figure 5: Young's modulus at varying temperatures for directionally solidified eutectic ceramic $MgAl_2O_4$-$Y_3Al_5O_{12}$.

measured to be 328 ± 91 GPa for MAS-YAG crystal. Similar results were obtained using a finite element method (FEM) for Al_2O_3-YAG system [40]. The Young's modulus of MAS-YAG crystal decreases with temperature and remains 298 ± 62 GPa at 1,400 °C.

Fracture toughness

The fracture toughness of MAS-YAG sample was determined by means of three point bending test using SENB method. The effect of temperature on the fracture toughness of the MAS-YAG crystal is shown in Figure 6. It can be seen that the fracture toughness of MAS-YAG eutectic slightly decreases when the temperature changes from 25 to 1,200 °C in vacuum. The toughness of MAS-YAG was 3.92 MPa $m^{1/2}$ at room temperature, 3.84 MPa $m^{1/2}$ at 1,200 °C. The fracture toughness of MAS-YAG material is not affected from temperature variation as much as elasticity. Although it has been reported that fracture toughness increases when the temperature of cubic complex crystals increases, MAS-YAG eutectic structure is not been affected in this situation [36]. At traditional materials, the increasing of toughness as much as the increasing of temperature and the decreasing of elasticity was expected. The retention of toughness's value together with temperature has been to explain why MAS-YAG crystal is not deformed at high temperatures. This situation has been evaluated at the following sections in terms of microstructure.

Microstructure analysis

Figure 7 shows the scanning electron microscope (SEM) image of the cross-section fracture surface for the directionally solidified eutectic MAS-YAG crystal after creep test at various temperatures. EDS analysis indicated that the bright area was $Y_3Al_5O_{12}$ and the dark area was $MgAl_2O_4$. MAS-YAG crystals show no grain growth at three different temperatures. A very limited ductile deformation has been seen at the samples that are exposed to creep test at 1,600 °C and 1,700 °C.

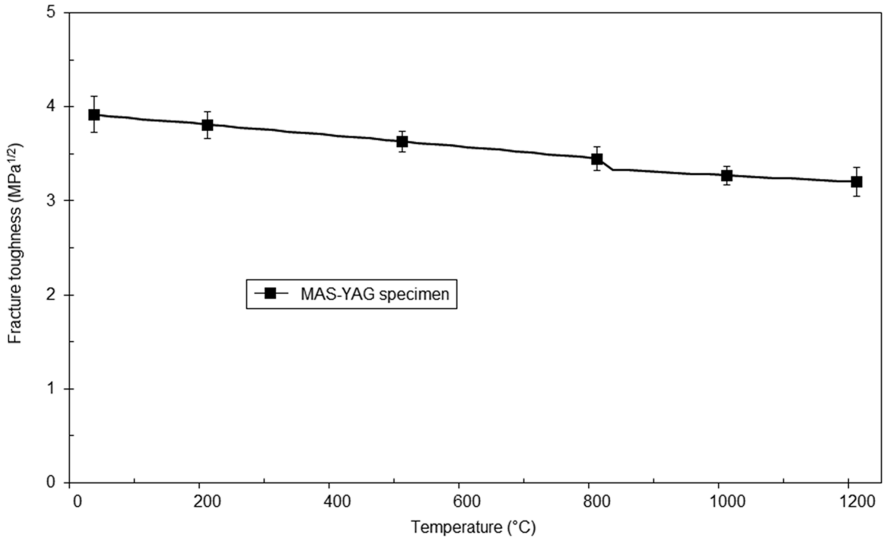

Figure 6: The fracture toughness of $MgAl_2O_4$-$Y_3Al_5O_{12}$ crystal as a function of temperatures.

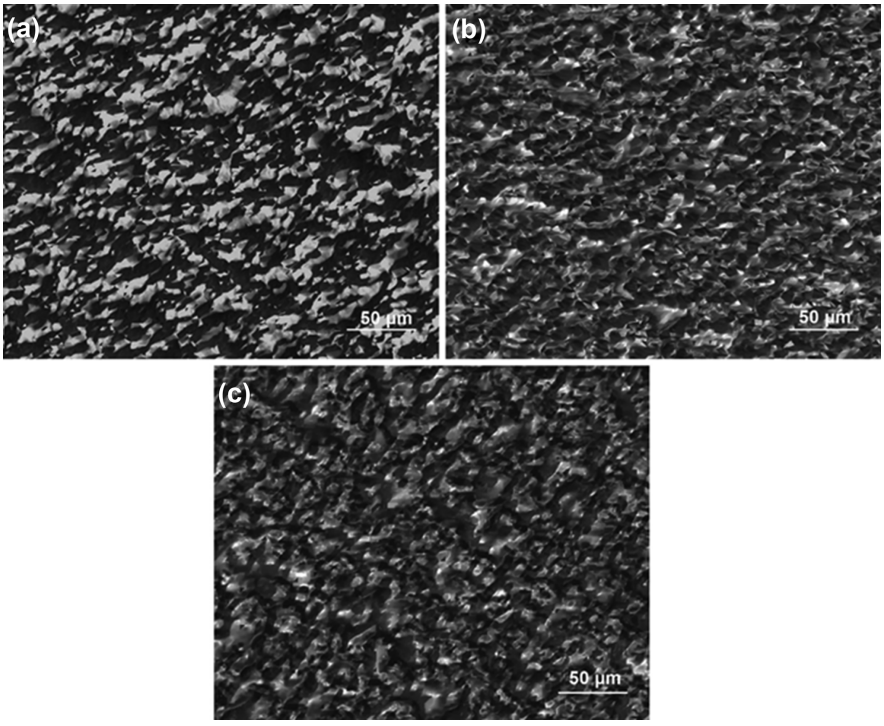

Figure 7: Scanning electron micrograph of the $MgAl_2O_4$-$Y_3Al_5O_{12}$ eutectic after creep deformation at various temperatures. For a directionally solidified MAS-YAG structure: (a) at 1,500 °C, (b) at 1,600 °C, and (c) at 1,700 °C. The bright areas are $Y_3Al_5O_{12}$ and the dark areas are $MgAl_2O_4$.

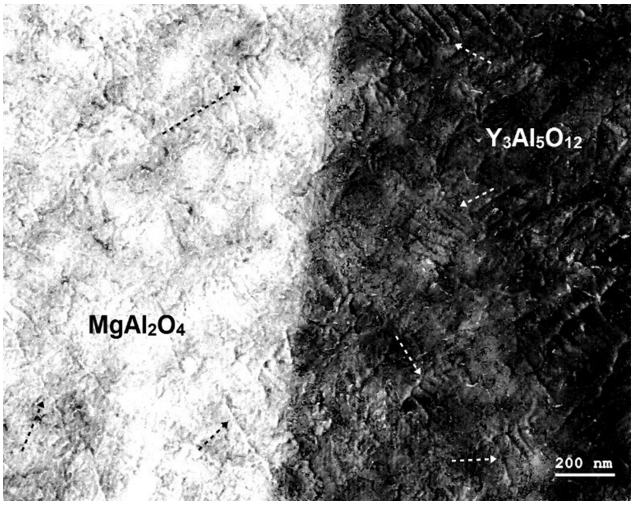

Figure 8: TEM images showing dislocation structures of $MgAl_2O_4$-$Y_3Al_5O_{12}$ phase boundary in the eutectic structure after the tensile test at 1,500 °C.

Figure 8 shows a TEM image of the microstructure of the plastically deformed DSMY specimen after the tensile test at 1,500 °C. Dislocation structures have been seen in both phases too. Formed plastic deformation is the result of dislocation motion. A plastic behavior occurring as a result of grain boundary sliding as it happened at polycrystals is out of question. The difference at deformation behaviors in both phases is due to discrepancies of creep resistances of both phases [41]. This discrepancy has obstructed seeing dislocation structure between two-phase boundary. Figure 9 shows SEM images of the fracture surface after tensile testing at 1,200 °C and 1,500 °C. The cracks shown in Figure 9(b) are to decrease tensile strength of overall structure when Figure 4 is considered. In this context, also no formation of cracks in Figure 9(a) is due to this reason. Despite the increase in the temperature, preservation of rigid structure explains why temperature has no impact upon toughness at directionally solidified eutectic MAS-YAG ceramics. As it has also been seen in Figure 8, it is possible that dislocations do not exist in the interface area and it is assumed that this situation provides that crack propagation stays in the structure.

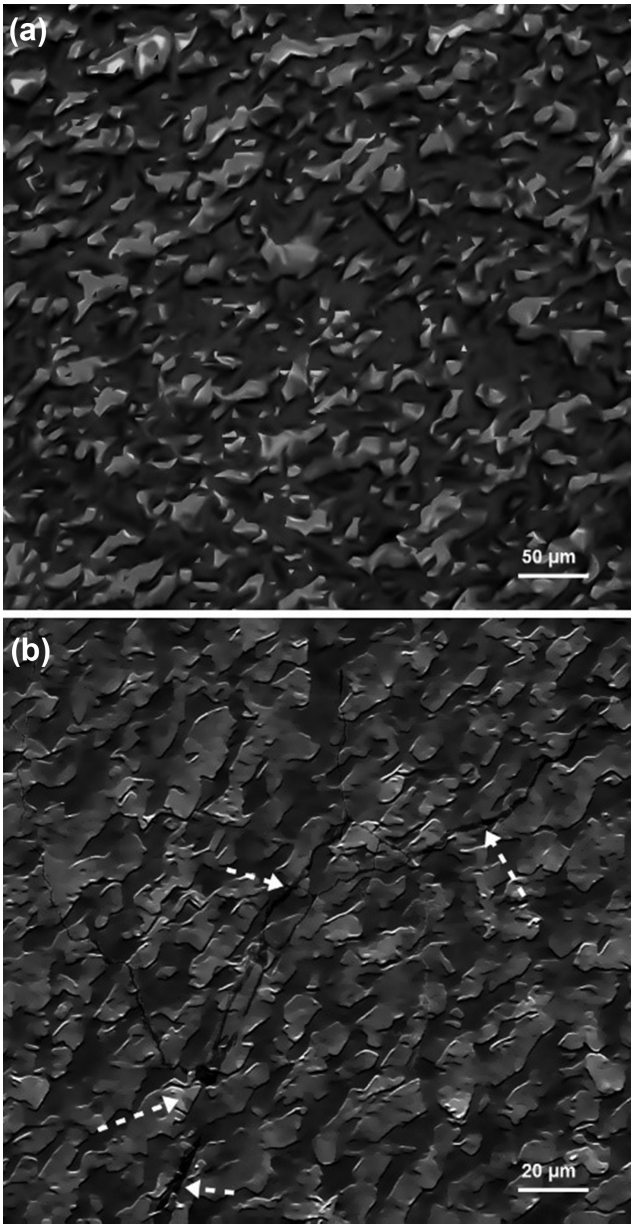

Figure 9: SEM images of the fracture surfaces for $MgAl_2O_4$-$Y_3Al_5O_{12}$ eutectics after the tensile tests (a) at 1,200 °C, and (b) at 1,500 °C. The bright areas are $Y_3Al_5O_{12}$ and the dark areas are $MgAl_2O_4$.

Conclusions

As well as temperature increase at MAS-YAG samples does not have too much effect on creep characteristics and toughness, especially after 1,200 °C, decreases at tensile strength and young modulus has been monitored. But these decreases are not different from the expected values for directionally solidified eutectic ceramics. Even though elasticity decreases, there does not exist much change at toughness value. Because the interface between MAS and YAG phases at the microstructure of MAS-YAG eutectic prevents dislocation mobility.

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