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Journal of the European Ceramic Society 25 (2005) 1307–1311

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Strain measurement of the directionally solidified eutectic $Al_2O_3/Y_3Al_5O_{12}$ (YAG) ceramic by neutron diffraction

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Abstract

The eutectic $Al_2O_3/Y_3Al_5O_{12}$ (YAG) ceramic has been reported to be composed by single-crystalline Al_2O_3 and YAG, the microstructure of which is characterized by the three dimensionally entangled two single-crystalline composites. Therefore, Laue diffraction and high-resolved energy-dispersive neutron diffraction (time-of-flight method) techniques were employed to measure residual strain precisely. It was found that the YAG phase was in tension and the Al_2O_3 phase was in compression with strains in the range of $\sim 10^{-4}$ at room temperature through comparing the lattice spacings of the sintered YAG and sintered Al_2O_3 as the references of strain-free materials.

Keywords: Composite; $Al_2O_3/Y_3Al_5O_{12}$ (YAG)

1. Introduction

The directionally solidified $Al_2O_3/Y_3Al_5O_{12}$ (YAG) eutectic single composite has attracted a lot of attention because of the superior flexural strength, thermal stability and creep resistance at high temperature.[1–7] A most striking structural characteristic is that the Al_2O_3/YAG composite consists of two phases of single-crystal Al_2O_3 and single-crystal YAG. The microstructure of this eutectic single composite observed by the scanning electron microscope (SEM) and transmission electron microscope (TEM) shows that the single-crystal Al_2O_3 and single-crystal YAG were interweaved three-dimensionally with each other. Mechanical and thermal characteristics that cannot be observed in the conventional ceramics are recognized to be strongly associated with the structure of the Al_2O_3/YAG eutectic single composite. Therefore, structural information at an atomic level is very important to know the origin of the outstanding mechanical properties.

In this paper, we report a crystallographic insight, especially the residual strain, of the directionally solidified Al_2O_3/YAG eutectic single composite for elucidating the high strength mechanism. The time-of-flight (TOF) Laue method of neutron diffraction using the high resolved neutron diffractometer *Sirius*[8,9] will give us a lot of structural information.

2. Experimental

Samples used in this study are directionally solidified eutectic single $Al_2O_3/Y_3Al_5O_{12}$ (YAG), sintered polycrystalline Al_2O_3 and $Y_3Al_5O_{12}$ (YAG). The eutectic single Al_2O_3/YAG was synthesized using the Bridgman-type equipment at the Japan Ultra-high Temperature Material Research Center. A mother ingot was obtained by preliminary melting of the mixed powders of high-purity commercial α-Al_2O_3 (AKP-30, Sumitomo Chemical Co. Ltd.) and Y_2O_3 (Y_2O_3-SU, Shin-etsu Chemical Co. Ltd.) powders. The ingot was inserted into a Mo-crucible situated in a vacuum chamber and

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doi:10.1016/j.jeurceramsoc.2005.01.026

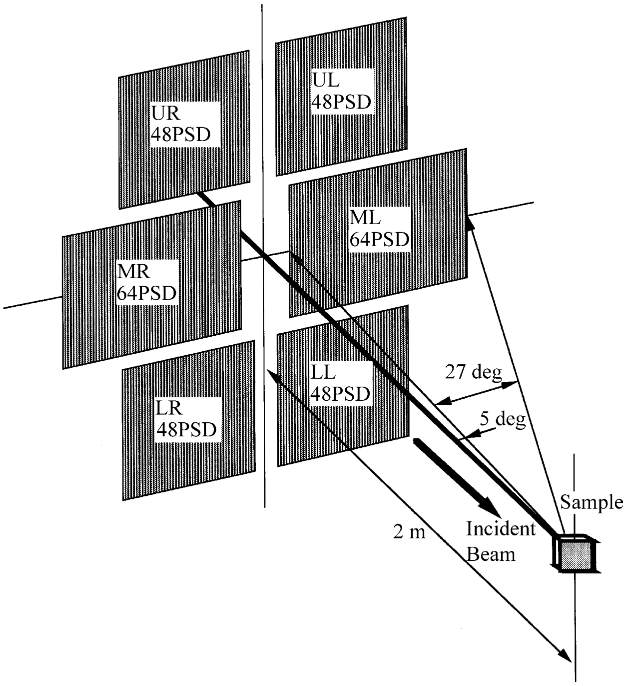

Fig. 1. Arrangement of six detector banks of *Sirius* backward bank; 64 one-dimensional position-sensitive detectors (PSDs) in the middle left (ML) and middle right (MR) banks and 48 in the lower left (LL), lower right (LR), upper left (UL) and upper right (UR) banks have been installed.

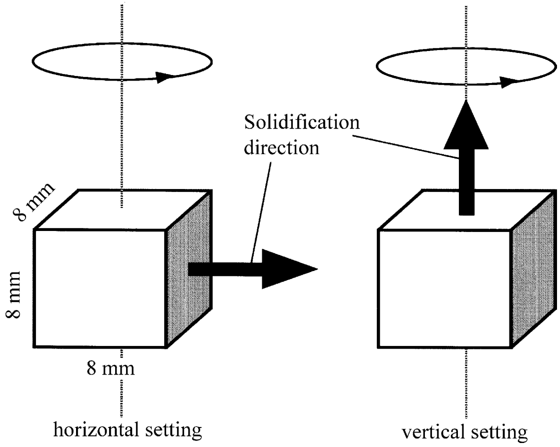

Fig. 2. Relationship between two settings and the solidification direction of the $Al_2O_3/Y_3Al_5O_{12}$ (YAG) composite.

then heated by a induction heating. After sustaining the melt at 2223 K for 30 min, the Mo crucible was lower at 5 mm/h to produce the unidirectional sample. Concerning the reference samples, the mixed powders obtained by ball milling were hot pressed in a carbon die to fabricate sintered samples at 1973 K under 50 MPa for an hour in a vacuum (10^{-2} mmHg). The detailed procedure of the sample preparation has been reported elsewhere.[3–7] Single-crystalline and sintered samples were cut into a cube with the edge dimension of 8 mm.

Residual strain measurements of the eutectic composite Al_2O_3/YAG were carried out with TOF Laue method using the neutron diffractometer *Sirius*[8,9] at Neutron Science Laboratory of High Energy Accelerator Organization (KEK) in Japan. We used the backward bank of *Sirius*, in which 320 one-dimensional position-sensitive detectors (PSDs) have been installed; six detector banks for backward scattering are situated 2 m apart from the sample position as shown in Fig. 1. The scattering angle in horizontal plane is $2\theta = 153°–175°$. The number of position pixels amounts to over 80,000. The TOF diffraction pattern can be obtained over wide θ range and wavelength ($\lambda = 0–0.5$ nm).

The single-crystalline Al_2O_3/YAG composite was placed in the two directions at the sample position of the *Sirius* diffractometer. The solidification direction was neatly aligned horizontally or vertically toward the neutron beam direction as shown in Fig. 2. A series of TOF Laue patterns were measured for about 150 min by each 12° rotating around the vertical axis. Fig. 3(A) and (B) shows the TOF Laue patterns of the samples which were set horizontally and vertically. In order to estimate the precise residual strain we focused on a selected Laue spot and summed up all TOF pattern in the detector region. The residual strain was evaluated by comparison of Bragg peaks of the Al_2O_3/YAG composite with corresponding peaks of the strain-free polycrystalline Al_2O_3 and YAG. The least-square curve fitting of the Bragg peaks was carried out to obtain the precise value of the lattice spacing.

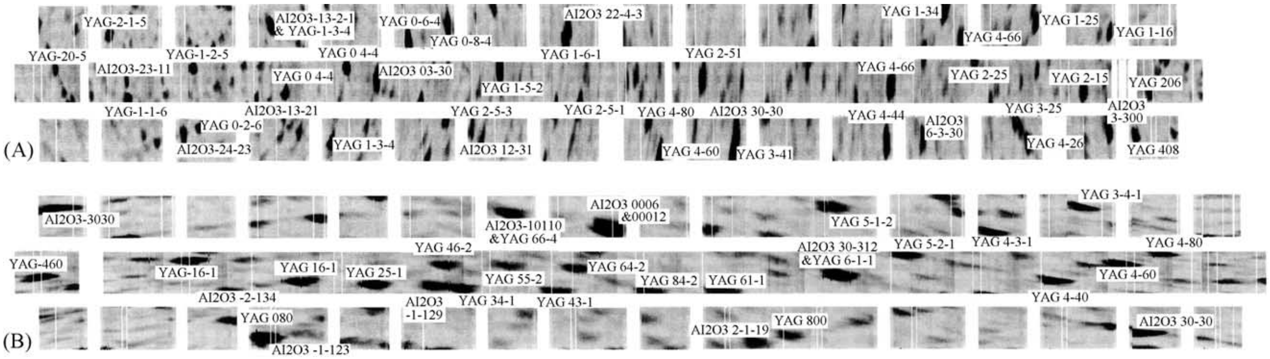

Fig. 3. Laue patterns of the horizontal setting (A) and vertical setting (B) samples toward the solidification direction for the $Al_2O_3/Y_3Al_5O_{12}$ (YAG) composite.

The residual strain measurement with TOF Laue method is based on the Bragg's law:

$$2d\sin\theta = \lambda, \tag{1}$$

where λ is neutron wavelength, d, lattice spacing and θ, scattering angle. When d changes from d^0_{hkl} to d_{hkl} elastically, the residual strain quantity ε_{hkl} is defined as follows;

$$\varepsilon_{hkl} = \frac{d_{hkl} - d^0_{hkl}}{d^0_{hkl}}, \tag{2}$$

where d^0_{hkl} is d-spacing without strain. The value of d^0_{hkl} represents a lattice spacing obtained from the strain-free polycrystalline samples.

All reflections were plotted on a Wulff net and stereographic projection to determine the reflection indices. The high-resolved TOF diffraction data obtained by *Sirius* spectrometer facilitate to index the reflections in the Laue pattern and give a precise lattice spacing of eutectic composite materials for evaluating the residual strain.

3. Results and discussion

Fig. 3(A) and (B) show all Laue diffraction patterns when the solidification direction of the sample was set horizontally and vertically toward the neutron beam direction. More than 150 Laue spots were confirmed to result from single-crystalline Al_2O_3 and YAG phases of the Al_2O_3/YAG composite. The reflections from $\{1\,1\,6\}$, $\{1\,3\,4\}$ and $\{1\,2\,5\}$ planes of the YAG phase were observed. In other Laue spots of the YAG phase, the $4\,\bar{8}\,0$, $4\,\bar{6}\,0$, $4\,\bar{4}\,0$ and $8\,0\,0$ reflections belong to the $[0\,0\,1]$ zone. Simultaneously, the $\bar{3}\,0\,3\,0$ and $3\,0\,\bar{3}\,0$ Laue spots of the Al_2O_3 phase were also observed. A Wulff net and a stereographic projection figure allow us to find that the YAG phase in the single Al_2O_3/YAG composite solidified along the direction close to $\bar{1}\,\bar{1}\,\bar{6}$ and that the Al_2O_3 phase to $\bar{2}\,3\,\bar{1}\,1$ in this sample. The solidification direction observed in this work is much different with those previously reported in the papers.[10,11] Therefore, we can say that the solidification directions of the YAG and Al_2O_3 phases in the single Al_2O_3/YAG composite depend on the solidification condition for the preparation.

The solidification direction is seen in Fig. 3(A). Laue spots near the solidification direction (left part of the figure) are sharp and isotropic. On the contrary, the Laue spots away from the solidification direction become broader. For example, the Laue spot of the $\bar{1}\,\bar{2}\,\bar{5}$ reflection of the YAG phase is circular with 3° × 2°, whereas that of the $2\,\bar{5}\,1$ reflection is oblong with 8° × 2°. In Fig. 3(B) where the solidification direction is vertical, all Laue spots spread out horizontally to be oblong with 3° × 5°–8°. These results suggest that when both Al_2O_3 and YAG phases in the Al_2O_3/YAG composite are tangled around the solidification direction, the reflection

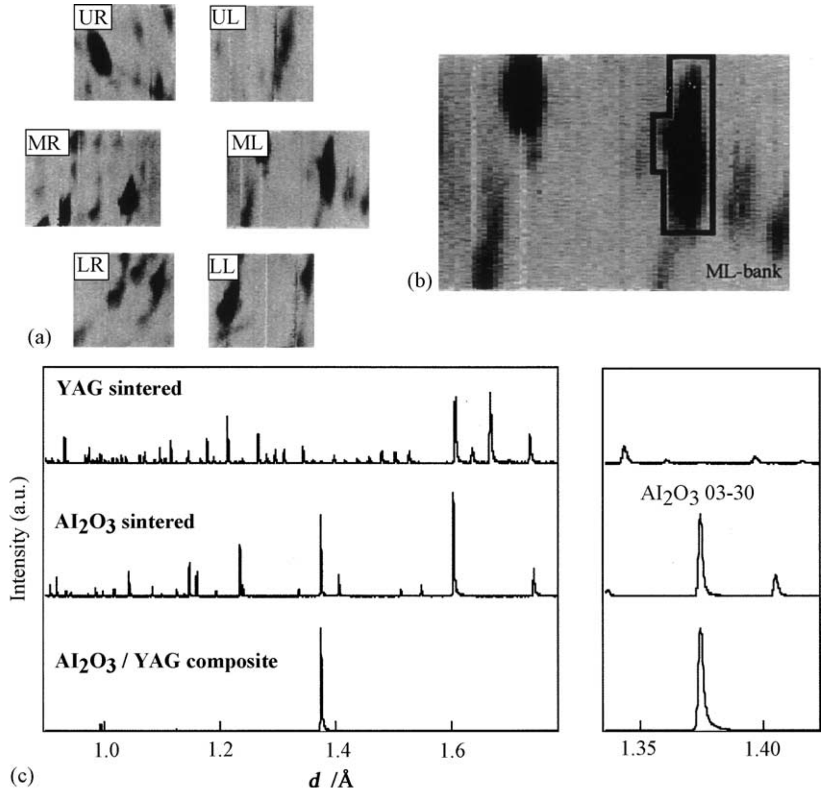

Fig. 4. Laue patterns observed by six backward detector banks: (a) and the middle left (ML) bank; (b) for the $Al_2O_3/Y_3Al_5O_{12}$ (YAG) composite, and the TOF data from the detectors surrounded by a solid line in the middle left (ML) bank; and (c) for the sintered Al_2O_3, sintered YAG and $Al_2O_3/Y_3Al_5O_{12}$ (YAG) composite.

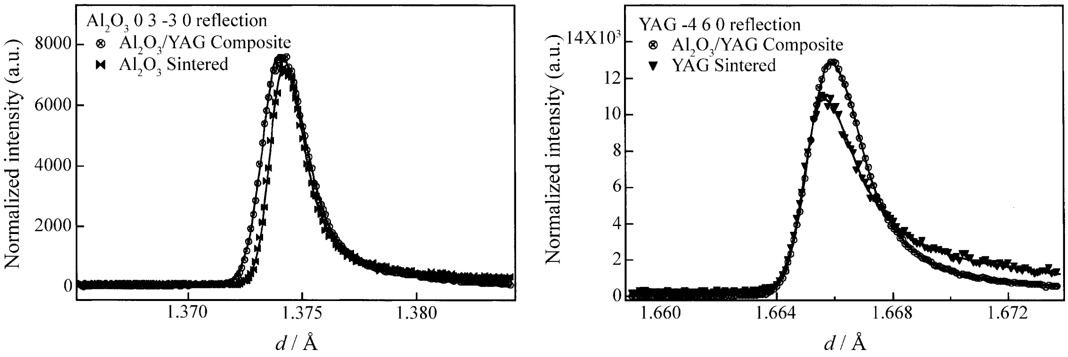

Fig. 5. Bragg peaks of Al_2O_3 $0\,3\,\bar{3}\,0$ and YAG $\bar{4}\,6\,0$ reflections for the sintered Al_2O_3, sintered YAG and $Al_2O_3/Y_3Al_5O_{12}$ (YAG) composite.

Table 1
The *d*-spacing and ε_{hkl} for the sintered Al_2O_3, sintered YAG and $Al_2O_3/Y_3Al_5O_{12}$ (YAG) composite

Reflection index *hkl*	*d*-spacing (nm)			$\varepsilon_{hkl}(\times 10^{-4})$
	$A1_2O_3$/YAG composite	$A1_2O_3$ sintered	YAG sintered	
$A1_2O_3$ phase				
$0\,3\,\bar{3}\,0$	0.137403	0.137419		−1.2
$\bar{2}\,\bar{1}\,3\,4$	0.140497	0.140519		−1.6
$0\,0\,0\,6$	0.216586	0.216662		−3.5
$2\,\bar{1}\,\bar{1}\,9$	0.123468	0.123504		−2.9
$3\,0\,\bar{3}\,1\,2$	0.085038	0.085069		−3.7
YAG phase				
$0\,\bar{6}\,\bar{4}$	0.166601		0.166538	3.8
$1\,\bar{6}\,\bar{1}$	0.194898		0.194835	3.2
$2\,\bar{5}\,1$	0.219379		0.219284	4.3
$4\,\bar{4}\,4$	0.173462		0.173394	3.9
$\bar{4}\,6\,0$	0.166585		0.166562	1.4

planes located away from the solidification direction in the reciprocal space are slightly tilted from the normal position to reduce the strain which results from the unidirectional solidification of two phases.

The evaluation of the residual strain was carried out by summing up all TOF patterns (Fig. 4(c)) in the detector region from the selected Laue spot (Fig. 4(b)) in the TOF Laue diffractometry for not only the Al_2O_3/YAG composite but also the polycrystalline Al_2O_3 and YAG, as shown in Fig. 4. The Bragg reflections of the polycrystalline Al_2O_3 and YAG in the corresponding detector region of the Al_2O_3/YAG composite were measured as the reference values of strain-free samples indispensably to get precise residual strains. Fig. 5 shows the enlarged Bragg peaks of Al_2O_3 $0\,3\,\bar{3}\,0$ reflection and YAG $\bar{4}\,6\,0$ reflection for the Al_2O_3/YAG composite, the polycrystalline Al_2O_3 and YAG, respectively.

Some of the residual strain values ε_{hkl} are summarized in Table 1. In both phases of the Al_2O_3/YAG composite the magnitude of ε_{hkl} was the order of 10^{-4} percent strain. Accordingly, it is quite small to measure appreciable residual strain within the measurement accuracy by a normal X-ray diffraction technique.[11] The strain value of the Al_2O_3/YAG composite is 10 times smaller than those of another composite samples.[11,12] Moreover, it is very important that all values of ε_{hkl} in the Al_2O_3 phase were found to be negative while those in the YAG phase positive. Same results have been reported in the previous measurement of the different sample with the dimensions of 5 mm × 5 mm × 50 mm. The order of magnitude of ε_{hkl} in the previous data[10,13] was also 10^{-4}. The signs of the residual strain of the Al_2O_3 and YAG phases indicate that the Al_2O_3 phase is in the compression state and the YAG phase is in the tension state. The structural characteristics that the sign of the strain value is negative and positive for the Al_2O_3 and YAG phases, respectively, and the orders are almost the same and small values of 10^{-4} may represent the causes of the high strength of the eutectic single Al_2O_3/YAG composite. Accordingly, in the future we should measure the variation of the residual strain of the eutectic single Al_2O_3/YAG composite depending on the temperature to clarify the mechanism of the high temperature strength and ductility.

4. Conclusions

The structural information obtained from the eutectic single $Al_2O_3/Y_3Al_5O_{12}$ (YAG) composite leads us to get the following conclusions.

(1) The TOF Laue method using the *Sirius* spectrometer allows us to determine the orientations of the Al_2O_3 and YAG phases of the directionally solidified Al_2O_3/YAG

composite. The YAG phase solidified along the direction close to $\bar{1}\,\bar{1}\,\bar{6}$ and the Al_2O_3 phase to $\bar{2}\,3\,\bar{1}\,1$, respectively.

(2) It is inferred from the horizontal broadening observed in the vertical setting of the sample that both single-crystalline Al_2O_3 and YAG phases were slightly distorted to reduce the structural strain resulted from the unidirectional solidification of the two phases.

(3) The high resolved measurement of the structure clearly indicates that the Al_2O_3 phase is in compression and the YAG phase is in tension with strains of the order of 10^{-4}.

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