

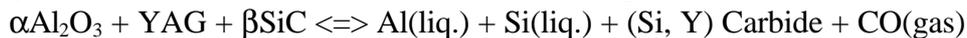
PREPARATION OF SiC REINFORCED ALUMINA-YAG EUTECTIC COMPOSITES

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SUMMARY: As a potential material for high temperature services, the mechanical properties and microstructure of the directionally solidified oxide eutectic alloys have been investigated. Currently, Waku et al. fabricated D.S. Al₂O₃-YAG eutectic composites. In this study, to improve the fracture toughness of this composites, the test specimens of SiC whisker or fiber / Al₂O₃-YAG composites were prepared by arc-melting or high frequency melting. In order to determine the chemical reaction between βSiC and eutectic liquid phase, EPMA, XRD and MDG analyses were carried out. As a results, the following reaction was estimated.



To prevent βSiC from oxidization, it is found that the pressure of CO gas should be larger than 4.43 atm at 2373 K. On the mechanical properties of the composites, micro-Vickers hardness, Hv of the Al₂O₃-YAG and the SiCw/ Al₂O₃-YAG composites are 1450 and 1680 respectively. The estimated fracture toughness K_{1C} of the composites by the indentation fracture method was approximately 9 MPa.m^{0.5}.

KEYWORDS: ceramics composite, SiC, alumina, YAG, eutectic, solidification

INTRODUCTION

In order to develop the materials for high temperature services, ceramics matrix composites have been studied. Currently, Waku et al. [1] fabricated directionally solidified Al₂O₃-YAG eutectic composites. As an advantage of this composites, the constituent phases are thermodynamically stable up to the eutectic temperature, 2109 K. Furthermore, the flexural strength of the composites keeps constant value from room temperature to 1973 K. Although this material reveals excellent properties in high temperature, the fracture toughness K_{1C}, 4 MPa.m^{0.5} at room temperature seems to be insufficient for practical use such as turbine blades of gas turbine engines. The final aims of this study are to improve the fracture toughness and to further improve high temperature properties of the Al₂O₃-YAG composites. Firstly, it is necessary to examine the chemical reaction between βSiC and eutectic liquid for the production of the composites during the solidification process. For this purpose, the test specimens of SiC/ Al₂O₃-YAG composites were prepared by arc-melting or high frequency heating. To determine the products of the reaction, EPMA, XRD and MDG analyses were carried out. On the mechanical properties of the composites, micro-Vickers hardness, Hv of the Al₂O₃-YAG and the SiCw/ Al₂O₃-YAG composites are measured. The fracture toughness of the composites was also estimated by the indentation fracture method.

EXPERIMENTAL PROCEDURE

Estimation of Chemical Reaction and Fracture Toughness

As starting materials, Al₂O₃ powder (AKP-30, Sumitomo Chemical Co., Ltd.), Y₂O₃ powder (Shin-Etsu Chemical Co., Ltd.) and βSiC whisker (TWS-400, Tokai Carbon Co., Ltd.) were used. The average dimensions of the whisker were a diameter of 10 μm and the length of about 500 μm. The oxide powders were dispersed in the ion exchanged water and mixed in the composition of Al₂O₃-18.5 mass% Y₂O₃, which is equal to the Al₂O₃-YAG eutectic composition. Then 40 mass% for the total weight of the oxide powders was added to the slurry. The slurry was slip-cast into the container to filtrate the water. After drying up the slurry, the mixture was melted in an arc-melting furnace. The solidified bulk was cut and polished. An optical microscope observation, EPMA, XRD and MDG (micro diffraction goniometry, collimator diameter : 100 μm) analyses were carried out in order to identify the products resulted from the chemical reaction. To estimate fracture toughness, K_{1C}, of the obtained material, indentation fracture method was applied. The crack length, which was introduced with micro-Vickers indentator at 9.8N, was observed with SEM. The toughness was approximately calculated by the crack length.

Trial Manufacture of SiC fiber/Al₂O₃-YAG Composites by "cast-in" Process

To confirm whether it is possible to produce the composites in the "cast-in" process, a sintered βSiC fiber (Tyran-SA, UBE Co. Ltd.) [2] and Al₂O₃-YAG eutectic alloy were used. Both the bundle of the fiber and the fragments of the oxide eutectic alloy were inserted into a molybdenum crucible, which have 8 mm inner-diameter and 12 mm high. The specimen was melted at the temperature of 2123 K, just over the eutectic temperature, in the vacuum of 1 Pa. The temperature was measured by a two color thermometer. The power of the heater was cut after keeping about 10 seconds at the temperature. The sample was polished and observed with an optical microscope.

RESULTS AND DISCUSSION

Estimation of the Chemical Reaction

Figure 1 shows the optical microscope and SEM images of the materials after arc-melting. Fig. 1(a) is a OM image of a material without SiC whisker. The composition of the starting material is the Al₂O₃-YAG eutectic, however primary YAG crystals was found. It may be caused by macro-segregation of YAG phase during the arc-melting. The microstructure with SiC whisker is shown in Fig. 1(b). It is found that the SiC whiskers are uniformly dispersed in the oxide matrix. The reaction products with metallic gloss are also sporadically found in the matrix. Fig. 1(c) shows a SEM image of a part of Fig. 1(b). If the sample was arc-melted continuously, the whisker and oxide materials were gradually consumed just under the argon arc-flame with production of gas phase. However, as to the remained whisker in the matrix, the change of the shape after arc-melting was scarcely found. The reaction products found in Fig. 1(b) are shown in Figure 2. As shown in this optical microscope image, the products resulted from the chemical reaction between SiC and the molten oxide are composed with at least three phases. During the solidification process of the products after arc-melting, the white faceted phase (phase-1) firstly crystallized in the melt of the products. Then gray faceted phase (phase-2) nucleated on the phase-1. Because the shape of phase-2 is allotromorphism to the shape of phase-1. The remained liquid (phase-3) solidified between phase-1 and phase-2. In order to

identify these phases, EPMA point analysis and MDG analysis were carried out. The points of the compositional analysis and the results of the semi-quantitative analysis were shown in Figure-3 and Table 1~3. The phase-1 consists of over 50 atomic % of carbon, silicon, yttrium and the other elements. The phase-2 will be silicon phase, whose melting point is 1703 K. The composition of the phase-3 is nearly equal to the Al-Si eutectic alloy (eutectic temperature, 850 K). These compositional analyses reveal that the phase-1 is a solid solution of silicon carbide including yttrium, and its melting temperature must be higher than the temperature of silicon phase. Figure 4 shows the profile of the MDG analysis on the same products. The collimator of diameter, 100 μ m, was used to adjust Cu-K α X-ray beam within an area of the products in a specimen. Aluminium and silicon phases were clearly detected from this profile. No candidate which fits the other unknown peaks was found in the ICDD cards. However, yttrium carbide or its solid solution will be a most possible phase found in the cards.

Table 1 Chemical composition of Phase 1.

Element	atomic %
Y	10.9
Al	8.0
Si	18.5
C	58.6
O	4.0

Table 2 Chemical composition of Phase 2.

Element	atomic %
Al	0.3
Si	99.7

Table 3 Chemical composition of Phase 3.

Element	atomic %
Al	83.0
Si	17.0

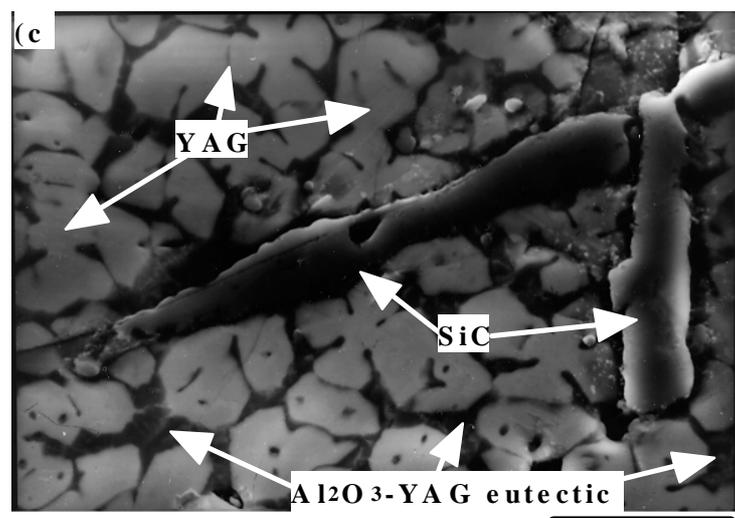
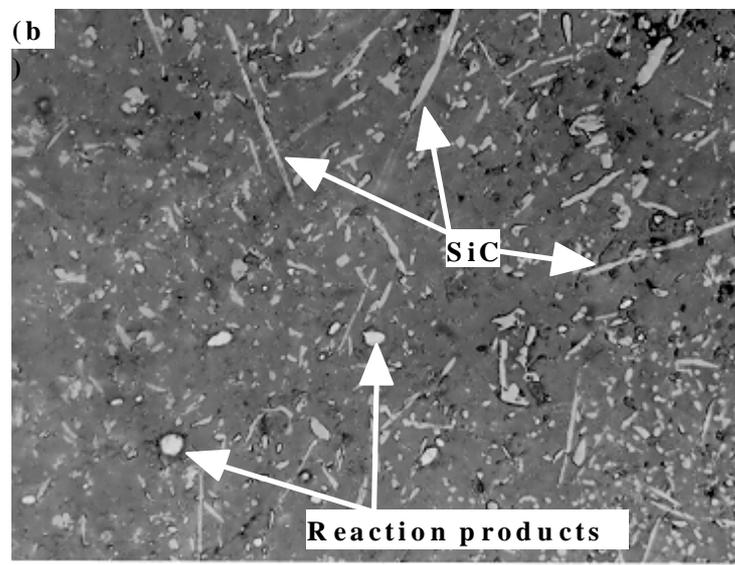
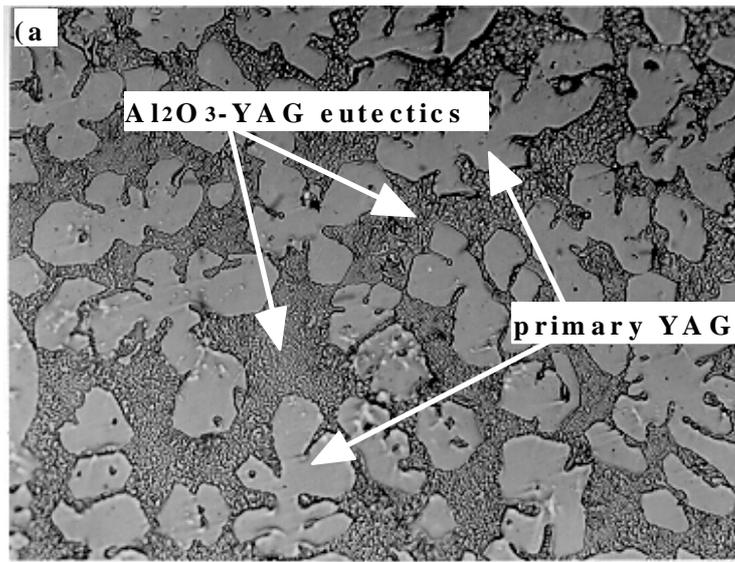


Fig. 1 (a) OM image of arc-melted Al_2O_3 -YAG eutectic alloy and (b) arc-melted SiC/ Al_2O_3 -YAG mixture, (c) SEM image of (b).

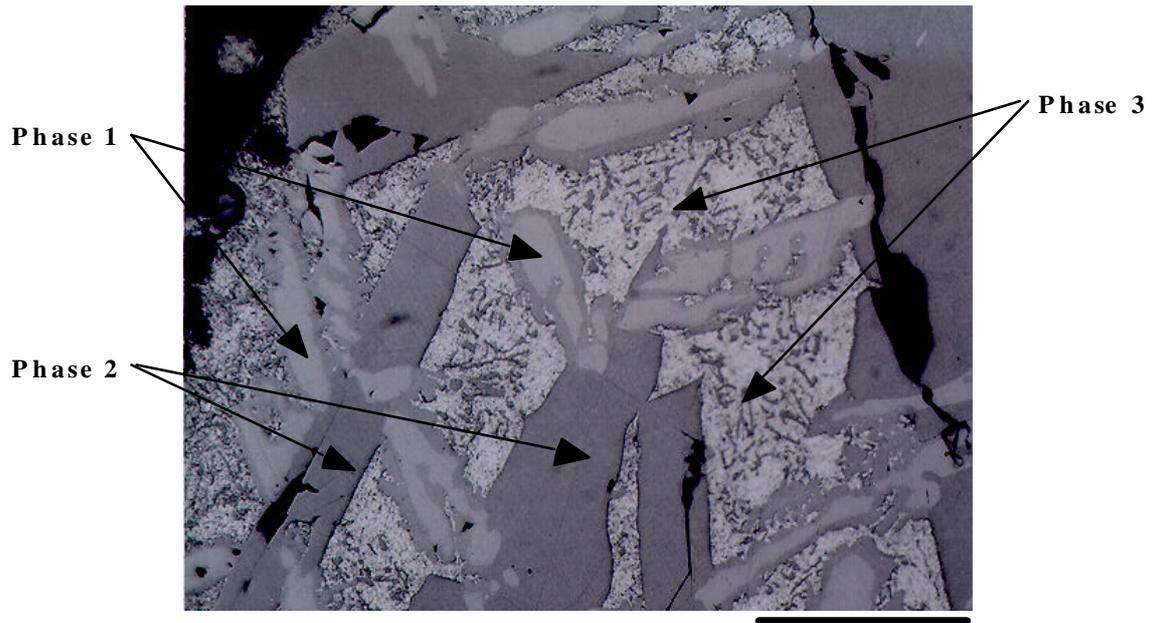


Fig. 2 OM image of reaction products in the Al₂O₃-YAG/SiC composite.

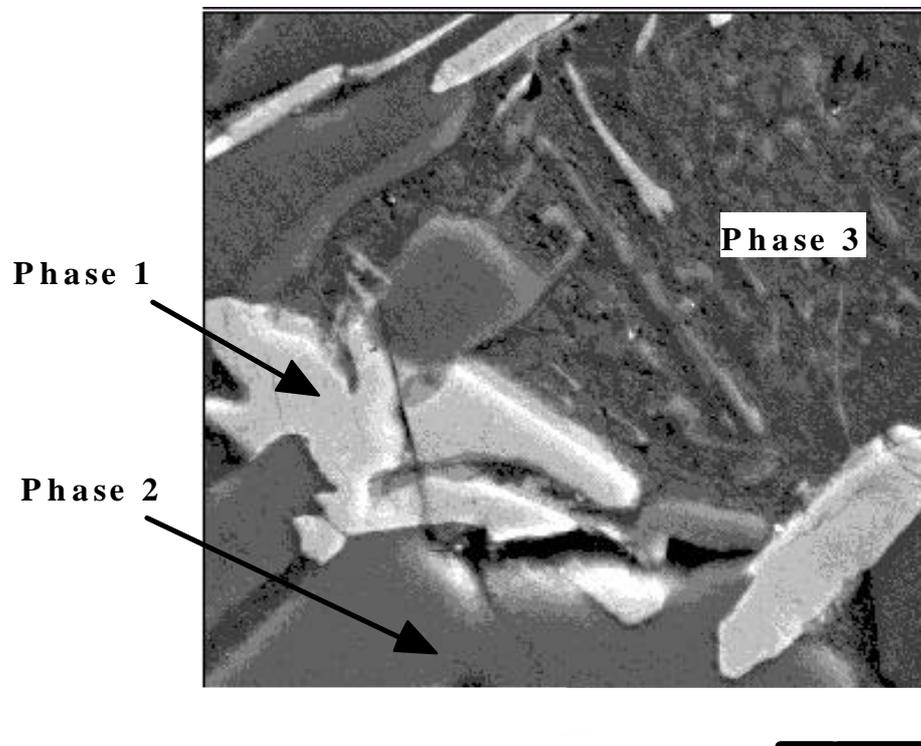


Fig. 3 SEM image of reaction products.

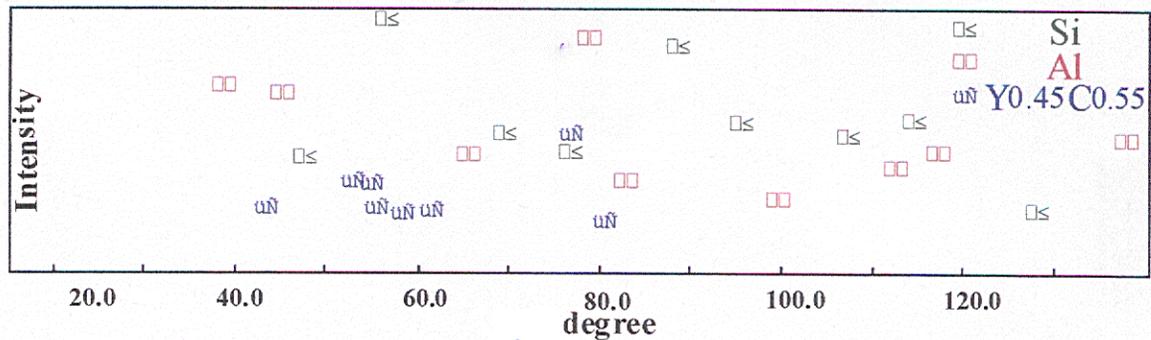
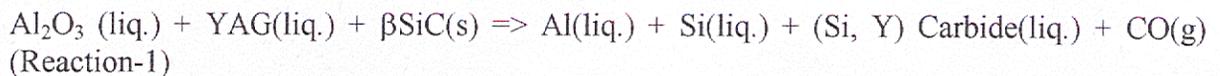


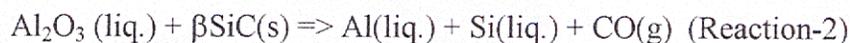
Fig. 4 MDG profile of reaction products in the Al₂O₃-YAG/ SiC composite.

Necessary Condition to Prevent the Chemical Reaction

From these analysis to determine the phases of products, the chemical reaction will be expressed as follows.



To prevent the reaction, it is necessary to examine Gibbs free energy change. However, as mentioned above, the carbide was not identified in the ICDD cards, and no thermodynamical data on the YAG phase was not found. For this reason, instead of the Reaction-1, Gibbs free energy of the following reaction, Reaction-2, was examined as a first approximation. The possibility of this reaction has also suggested by M. A. Mulla et al. [3]



The equilibrium CO gas pressure of the Reaction-2 should be solved in order to examine the necessary condition to produce the composites without chemical reaction [4][5]. The temperature to calculate the energy was to be set at over the eutectic temperature, 2099 K. In this case the assumed temperature in the Reaction-2 was 2373 K, which was higher than the melting point of alumina, 2325 K. The following are the equation represents the equilibrium, and the other assumptions.

$$\Delta G = \Delta G_0 + RT \ln K = 0, \text{ at } 2373\text{K (Eq. 2)}$$

$$a_{\text{Al}_2\text{O}_3 (\text{liq.})} = a_{\text{SiC}(\text{s})} = 1$$

$$a_{\text{Al}} = 0.83, a_{\text{Si}} = 0.17 \text{ (ideal solution)}$$

As a result, it is found that at least 4.43 atm of partial CO pressure is necessary to prevent liquid alumina from the deoxidation by silicon carbide.

Estimation of the Fracture Toughness

Figure 5 shows the SEM images of the Al₂O₃-YAG and SiC/ Al₂O₃-YAG composites after the measurement of micro-Vickers hardness. The hardness, average crack length and estimated K_{1C} are shown in Table 4. The K_{1C} is increased with the addition of the whisker, however,

K_{1C} of the SiC/ Al_2O_3 -YAG is not necessarily reliable. Because most of cracks does not correspond to typical median crack, which grows straight from the indents.

Table 4 micro-Vickers hardness and fracture toughness K_{1C} of the composites.

	Al_2O_3 -YAG	Al_2O_3 -YAG
Hv	1450	1680
Av. Crack length, μm	85	45
K_{1C} , $MPa \cdot m^{0.5}$	4 [6]	8.7

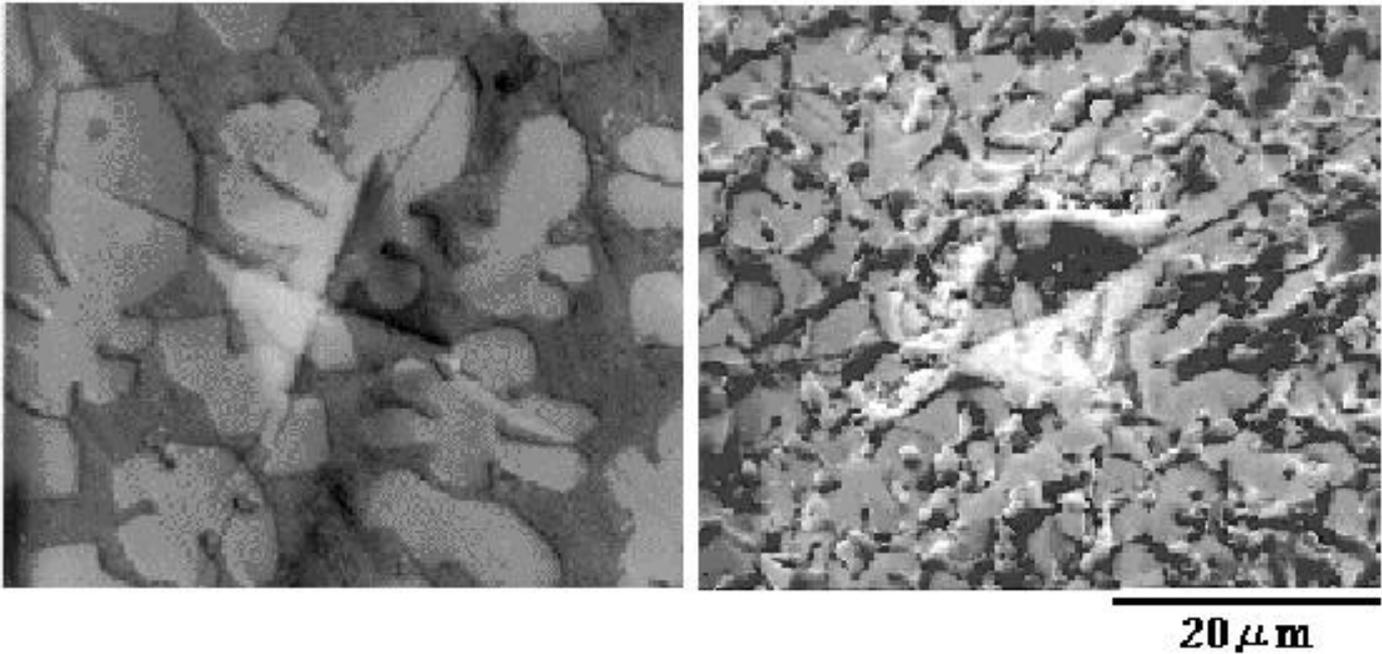


Fig. 5 SEM images of Al_2O_3 -YAG and SiC w/ Al_2O_3 -YAG materials after indentation

Trial Manufacture of SiC fiber/ Al_2O_3 -YAG Composites by "cast-in" Process

Figure 6 shows the optical microscope image of the SiC/ Al_2O_3 -YAG composites. The SiC fiber set in a molybdenum crucible with the fragments of Al_2O_3 -YAG eutectic alloy was heated up to 2123 K and kept for about 10 seconds under the pressure of 1Pa. The introduction of the CO pressure into the chamber of the furnace was avoided from the view point of the safety of the experiment. However, as shown in this figure, the fibers were included by the eutectic oxides and the reaction products were scarcely found in the specimen. In this experiment, the bundle of the fibers and fragments of the oxide were separately located in the crucible. Therefore it is clear that the molten oxide spontaneously infiltrate into the bundle of the SiC fibers and this made the composites microstructure. This maybe caused by the excellent wettability between surface SiO_2 film of the fiber and the molten eutectic oxide. If the SiC fiber contacts with molten oxide for long time, the chemical reaction must be inevitable. Therefore, the fiber needs to be coated with a certain kind of prohibitive agency of the reaction, for example platinum or rhodium.

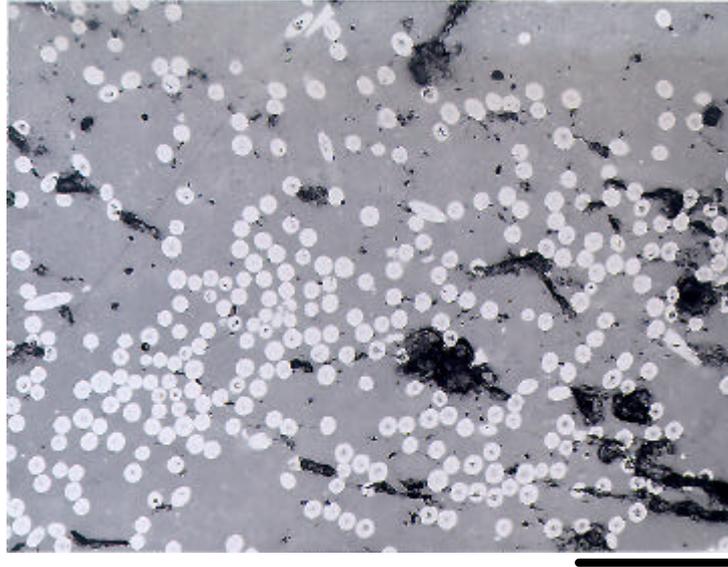
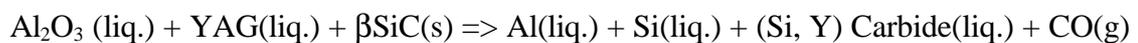


Fig. 6 OM image of SiCf/Al₂O₃ -YAG composite.

CONCLUSIONS

In order to prepare SiC whisker or fiber reinforced Al₂O₃-YAG eutectic composites by "cast-in" process, the chemical reaction between SiC and the molten oxides were examined.

- (1) After arc-melting or high frequency heating of the mixture of SiC and Al₂O₃-YAG eutectic alloy, the composites microstructure was obtained, however, the products resulted from the chemical reaction between SiC and molten alloy were also found in the oxides matrix.
- (2) The chemical reaction was determined as follows with EPMA and MDG. However, the carbide phase including silicon and yttrium was not identified in the ICDD card.



- (3) From a result of first approximation of Gibbs free energy, at least 4.43 atm of partial CO gas pressure is necessary at 2373 K to prevent the chemical reaction.

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