### Ceramic Processing Research

### Processing of SiC fiber/alumina-mullite matrix composites

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Yb-doped ceria with a composition of  $Ce_{0.8}Yb_{0.2}O_{1.9}$  was used to coat SiC fibers by a solution immersion route using  $Ce(NO_3)_3$ and Yb(NO<sub>3</sub>)<sub>3</sub>. The phase compatibility between the fibers and the coating layer was studied in an Ar atmosphere and in air at 1100°-1300°C. The heat-treatment of the mixture of Yb-doped ceria/SiC fibers with an Al sintering additive (<1 mass%) in an Ar atmosphere at 1300°C produced CeAlO<sub>3</sub> and 7Ce<sub>2</sub>O<sub>3</sub> · 9SiO<sub>2</sub>. Subsequent heating of the fibers in air at 1100°-1300°C decomposed the CeAlO<sub>3</sub> and 7Ce<sub>2</sub>O<sub>3</sub> · 9SiO<sub>2</sub> into CeO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub> and SiO<sub>2</sub>. The SiC fibers coated with Yb-doped ceria and 80 mass% mullite-20 mass% alumina sheets of 75  $\mu$ m thickness, were laminated and hot-pressed at 1200°C in an Ar atmosphere. The as hot-pressed composite with 15 vol% fibers and the composite subsequently heated in air at 1100°C showed ductile and brittle properties in the stress-strain relations, respectively. These results were associated with the phase stability at the interfaces between the fibers and matrix.

Key words: Yb-doped ceria, Interface, SiC fibers, Phase compatibility, Mullite-alumina matrix, Hot-pressing.

### Introduction

It is well known that continuous fiber-reinforced ceramic matrix composites with weak interfaces and small sliding resistance are effective in improving the performance of brittle ceramics to give a nonlinear stress-strain behavior and a large work of fracture which arises due to the interfacial debond and subsequent fiber pull out during crack propagation [1-2]. The successful mechanical properties of oxide fibers/ oxide matrix composites depend greatly upon the incorporation of interfacial material which is compatible with the debonding of fibers and the chemical stability to the fibers and oxide matrix. A carbon or BN layer with sufficient sliding characteristics as an interfacial material promotes the fiber pull out in the oxide fibers/ oxide matrix composites or nonoxide fiber-reinforced ceramic matrix composites. The use of those composites is limited in an atmosphere of low oxygen pressure because of the oxidation of the interfacial materials. For this reason, oxide coatings of ZrO<sub>2</sub> [3], LaPO<sub>4</sub> [4], and rare-earth hexa-aluminate [5, 6] have been investigated as interfacial materials in oxide fibers/ oxide matrix composites.

In a previous paper [7], we have studied the processing of ceria-coated alumina fibers/alumina matrix composite by hot-pressing at 1200°C. The grain growth in the alumina fibers and ceria layer increased the roughness at the interfaces and decreased the strength of the fibers, inducing brittle fracture of the composite. Based on this result, we intend to understand more about the suitable processing and resultant mechanical properties of oxide fibers/oxide matrix composites. The following change was introduced to the fiber, interphase and matrix of the ceria-coated alumina fibers/alumina matrix composite. It is difficult to control the properties of the as-received commercial oxide fibers at a high temperature. The alumina fibers were replaced by high strength SiC fibers (tensile strength 2.5 GPa) to suppress the grain growth and degradation during the processing. After the establishment of the processing conditions, the SiC fibers may be again changed to suitable oxide fibers with no degradation at high temperatures. According to the manufacture [8], the SiC fibers show no degradation in strength on heating to 1900°C in an inert atmosphere. The fibers were produced from amorphous Si-Al-C-O fibers by the release of CO gas at temperatures from 1500° to 1700°C and sintering at temperatures over 1800°C. The sintered SiC fiberbonded ceramics were also produced by hot-pressing piled sheets of the amorphous SiC fibers, which showed nonlinear stress-strain curves reaching 600 MPa in four-point flexural strength up to 1600°C in air [9]. When the alumina matrix with a  $8 \times 10^{-6}$  K<sup>-1</sup> thermal expansion coefficient (TEC) was hot-pressed with 20 vol% Si-C-O fibers with a  $3 \times 10^{-6}$  K<sup>-1</sup> TEC, many microcracks were formed in the fibers and matrix at room temperature owing to the large internal stresses resulting from the difference of Young's modulus and

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TEC between matrix and fibers [10]. The residual tensile stress induced in the alumina matrix ( $\sigma_m^*$ ) is calculated from Eq. (1) [11],

$$\sigma_{\rm m}^* = E_{\rm c} \varepsilon_{\rm m}^* = E_{\rm f} V_{\rm f} \Delta \alpha \Delta T \tag{1}$$

where  $E_c$  is the Young's modulus of the composite given by Eq. (2),  $\varepsilon_m^*$  the residual strain formed in the matrix at room temperature,  $E_f$  the Young's modulus of the fibers,  $V_f$  the volume fraction of the fibers,  $E_m$  the Young's modulus of the matrix,  $\Delta \alpha$ , the difference (=  $\alpha_m - \alpha_f$ ) of the TEC between the matrix ( $\alpha_m$ ) and the fiber ( $\alpha_f$ ), and  $\Delta T$  the difference between the processing temperature and room temperature.

$$E_{c} = E_{f}V_{f} + E_{m}(1-V_{f})$$
 (2)

The condition  $\varepsilon_m^* \ge \varepsilon_{um}$  ( $\varepsilon_{um}$ : critical strain in the matrix to produce fracture) results in fracture of matrix. The TEC of the matrix was decreased by adding mullite powder with a  $5 \times 10^{-6} \text{ K}^{-1}$  TEC to the alumina to avoid the formation of cracks in matrix after processing. Higashi et al. [12] reported that doping with a rareearth element (especially, a smaller element such as Yb) into ceria is effective in suppressing grain growth. In the present experiment, Yb-doped ceria was used to coat the sintered SiC fibers. As described above, the objective of the present work is to establish the processing conditions for dense laminated composites of sintered SiC fibers/Yb-doped ceria (interphase)/mullitealumina matrix system. The result obtained will be reflected in the next stage of the processing of oxide fibers/oxide interphase/oxide matrix systems.

### **Experimental Procedure**

#### Coating of SiC fibers with Yb-doped ceria

The properties of sintered SiC fibers supplied by Ube Industries Ltd., Japan are as follows: diameter 7.5 µm, density  $3.0 \text{ g/cm}^3$ , sintering additive Al < 1 mass%, Young's modulus 412 GPa, tensile strength 2.79 GPa and thermal expansion coefficient  $4 \times 10^{-6}$  K<sup>-1</sup>. The surface of the SiC fibers was coated with a Yb-doped ceria layer by immersion into a mixed aqueous solution containing 0.4 M-Ce(NO<sub>3</sub>) 3 and 0.1 M-Yb(NO<sub>3</sub>) 3 at pH 5.2. The following immiscible liquid displacement method was used to coat the surface of SiC fibers with Yb-doped ceria. The fibers were withdrawn slowly from the lower layer of a Ce-Yb nitrate solution through an upper layer of octanol. The excess aqueous solution on the surface of the fibers was washed down at the interface between the octanol and the aqueous solution due to the different surface energies of the liquids. The dipped fibers were heat-treated at 400°C in air for 20 minutes. This dipping process was repeated five times. The phase compatibility of the SiC fibers-Yb-doped ceria system and the Al<sub>2</sub>O<sub>3</sub>-SiO<sub>2</sub>-CeO<sub>2</sub> system was studied in an Ar atmosphere and in air at 1100°-1300 °C. The crushed SiC fibers, Ce(NO<sub>3</sub>)<sub>3</sub>-derived CeO<sub>2</sub>, and Yb(NO<sub>3</sub>)<sub>3</sub>-derived Yb<sub>2</sub>O<sub>3</sub> were mixed in the mass ratio of SiC fibers/CeO<sub>2</sub>/Yb<sub>2</sub>O<sub>3</sub> = 76.9/18.0/5.1. This ratio was based on the weight of fibers after 5 timesdipping in the mixed nitrate solution. The mixtures were heated at 1100°-1300°C for 2 h in an Ar atmosphere (99.99 vol%) or in air. A high purity  $\alpha$ -alumina powder (>99.99 mass%, AKP 50, Sumitomo Chemical Industry Co., Ltd., Tokyo, Japan), tetraethyl orthosilicate (Si(OC<sub>2</sub>H<sub>5</sub>)<sub>4</sub>)-derived silica powder and Ce(NO<sub>3</sub>)<sub>3</sub>derived CeO<sub>2</sub> powder were mixed in the mass ratio  $Al_2O_3/SiO_2/CeO_2 = 74.5/13.2/12.3$  and heated at 1100°-1300°C for 2 h in an Ar atmosphere and in air. The phases produced in the reducing and oxidizing atmosphere were studied by X-ray powder diffraction. The concentration gradient of Al, Si, Ce and Yb was also measured for the Yb-doped ceria-coated SiC fiber, and ceria-coated silica-alumina fibers heated in an Ar atmosphere and in air at 1100°C, by an electron probe microanalyzer (JSM-820, Joel Co., Ltd., Japan). The silica-alumina woven fabric (1000 filament/yarn) was made of long  $\gamma$ -alumina fibers 15  $\mu$ m in diameter with the chemical composition of 85 mass% Al<sub>2</sub>O<sub>3</sub> and 15 mass% SiO<sub>2</sub> (Sumitomo Chemical Co., Ltd., Japan). The silica-alumina fabric was dipped into 0.25 M-Ce(NO<sub>3</sub>)<sub>3</sub> solution at pH 6.0 and heated at 400°C for 20 minutes. This sequence was repeated four times.

### Preparation of mullite-alumina sheet by a doctor blade method

A sol-gel-processed crystalline mullite powder consisting of flaky particles (specific surface area of 5.7  $m^2/g$ , composition of 71.80 mass% Al<sub>2</sub>O<sub>3</sub>, 28.05 mass% SiO<sub>2</sub>, 0.30 mass% ZrO<sub>2</sub>, 0.10 mass% TiO<sub>2</sub>, 0.01 mass% Na<sub>2</sub>O, Chitibu Cement Ltd., Japan) and  $\alpha$ -alumina powder (impurity: Na 50 ppm, Fe 10 ppm, Si 10 ppm, Asahi Chemical Industry Co., Ltd., Japan) of specific surface area 8.0 m<sup>2</sup>/g (equivalent spherical diameter 0.19 µm) were mixed in the mass ratio of mullite/ alumina = 80/20 and then milled with Y<sub>2</sub>O<sub>3</sub>-partially stabilized ZrO<sub>2</sub> balls of 3 mm diameter for 72 h in a polyethylene container. The cumulative particle size was measured using a particle size analyzer by laser diffraction (Helos & Rodos, Nihon Denshi Co., Tokyo, Japan). An aqueous suspension of the mixed powders was prepared with 20 vol% solid, polyacrylic ammonium dispersant (PAA, average molecular weight 10000, Dai-ichi Kogyo Seiyaku Co., Ltd., Japan) of 0.6 mass%, and methyl cellulose binder (molecular weight 63000) of 2.0 mass% against the weight of solid. The suspension was formed on polyester sheets by a doctor blade method (DP-100, Tsugawa Seiki Seisakusho, Tokyo, Japan) at a transfer rate of 15 cm/minute. The front and back blade clearance was controlled to 300 and 400 μm, respectively, to form oxide sheets of 75 μm thickness and 49.3% theoretical density. The sheets were cut into 38 mm length and 25 mm width pieces to hotpress with the SiC fibers.

## Processing of SiC fibers coated with Yb-doped ceria/mullite-alumina matrix composite

The unidirectional SiC fibers coated with Yb-doped ceria were distributed on the mullite-alumina sheets and laminated. The laminated green composite with 15 vol% fibers was pressed using a carbon die under a pressure of 39 MPa at room temperature and heated to 1200°C at 10 K/minute, and sintered for 2 h in an Ar atmosphere (FVH-5 type, Fuji Dempa Kogyo Co., Japan). The density of the sintered composite was measured by the Archimedes method using distilled water. The hot-pressed composite was cut by a diamond cutting wheel into five test specimens 38 mm long, 4 mm wide, and 3 mm high and polished with 1 µm diamond paste. The flexural strength of the test specimens parallel to the hot-pressing direction were measured at room temperature by a four-point flexural method over spans of 10 mm (upper span) and 30 mm (lower span) at a cross-head speed of 0.5 mm/minute (Model UTM-1-5000 BE, Tokyo Baldwin Co., Tokyo, Japan). A stain gauge was attached to the tensile plane of the composite.

### **Results and Discussion**

### Chemical reaction between SiC fibers and Ybdoped ceria

Figure 1 shows the XRD patterns of the powder mixtures of  $Al_2O_3/SiO_2/CeO_2 = 74.5/13.2/12.3$ , heated at 1100°-1300°C in an Ar atmosphere for 2 h (a, b, c) and subsequently heated in air for 2 h (a', b', c'). In the alumina-silica-ceria system in air [3], alumina, mullite and ceria coexisted as the stable phases. The system in an Ar atmosphere produced CeAlO<sub>3</sub> and 7Ce<sub>2</sub>O<sub>3</sub> · 9SiO<sub>2</sub> below 1200°C, indicating the reduction of Ce<sup>4+</sup> ions. However, these compounds decomposed to amorphous phases at 1300°C and also crystallized into mullite, Al<sub>2</sub>O<sub>3</sub> and CeO<sub>2</sub> in air at 1300°C. Equations (3) and (4)



**Fig. 1.** XRD patterns for the powder mixtures of  $Al_2O_3/SiO_2/CeO_2 = 74.5/13.2/12.3$ , heated at 1100°-1300°C in an Ar atmosphere for 2 h (a, b, c) and subsequently heated in air for 2 h (a', b', c').

correspond to the chemical reaction of the aluminaceria system and the silica-ceria system, respectively. The lack of formation of mullite in air below 1200°C indicates the low diffusion rates of cations and the lack of a crystalline phase of silica at 1100°C in air indicates the formation of amorphous silica.

$$2\text{CeO}_2 + \text{Al}_2\text{O}_3 \xrightarrow{\text{Ar}} 2\text{CeAlO}_3 + \frac{1}{2}\text{O}_2$$
 (3)

$$14\text{CeO}_2 + 9\text{SiO}_2 \quad \underbrace{\text{Ar}}_{\text{Air}} \quad 7\text{Ce}_2\text{O}_3 \cdot 9\text{SiO}_2 + \frac{7}{2}\text{O}_2 \quad (4)$$

Figure 2 shows the crystalline phases of the mixtures of SiC fibers/CeO<sub>2</sub>/Yb<sub>2</sub>O<sub>3</sub> = 76.9/18.0/5.1, heated at 1100°-1300°C in an Ar atmosphere. In addition to SiC and CeO<sub>2</sub> ss, CeAlO<sub>3</sub> and 7Ce<sub>2</sub>O<sub>3</sub>·9SiO<sub>2</sub> were identified. Equations (5) and (6) show the possible reactions for the oxidation of SiC (Eq. (5)) and the formation of 7Ce<sub>2</sub>O<sub>3</sub>·9SiO<sub>2</sub> [13]. Equation (7) corresponds to the total reaction.

$$\operatorname{SiC} + \frac{15}{2} \operatorname{Ce}_{0.8} \operatorname{Yb}_{0.2} \operatorname{O}_{1.9} \longrightarrow$$
  
$$\operatorname{SiO}_2 + 3\operatorname{Ce}_2 \operatorname{O}_3 + \frac{3}{4} \operatorname{Yb}_2 \operatorname{O}_3 + \operatorname{CO} \uparrow \qquad (5)$$

$$7Ce_2O_3 + 9SiO_2 \longrightarrow 7Ce_2O_3 \cdot 9SiO_2$$
(6)

$$9\text{SiC} + \frac{135}{2} \text{ Ce}_{0.8}\text{Yb}_{0.2}\text{O}_{1.9} \longrightarrow 7\text{Ce}_2\text{O}_3 \cdot 9\text{SiO}_2 + 20\text{Ce}_2\text{O}_3 + \frac{27}{4}\text{Yb}_2\text{O}_3 + 9\text{CO} \uparrow (7)$$

The compound  $CeAlO_3$  was formed by the reaction between  $Ce_{0.8}Y_{0.2}O_{1.9}$  and the Al component contained in the SiC fibers. The formation process of  $CeAlO_3$ 



**Fig. 2.** XRD patterns for the powder mixtures of SiC fibers/CeO<sub>2</sub>/ $Yb_2O_3 = 76.9/18.0/15.1$ , heated at  $1100^{\circ}-1300^{\circ}C$  in an Ar atmosphere for 2 h (a, b, c) and subsequently heated in air for 2 h (a', b', c').

may follow Eq. (8).

$$2Al + Ce_2O_3 + 3CO \longrightarrow 2CeAlO_3 + 3C \qquad (8)$$

The significant difference between Eq. (7) and the result in Fig. 2 is that  $Ce_2O_3$  was not identified in the experiment, suggesting an amorphous state for  $Ce_2O_3$ . Furthermore, the compounds of CeAlO<sub>3</sub> and  $7Ce_2O_3$ . 9SiO<sub>2</sub> were stable under the coexistence of SiC at 1300 °C and this result was different from the result under the coexistence of Al<sub>2</sub>O<sub>3</sub> (Fig. 1). The disappearance of CeAlO<sub>3</sub> and  $7Ce_2O_3$ .9SiO<sub>2</sub> by subsequent heating in air is explained by the decomposition of these compounds and oxidation of Ce<sup>3+</sup> ions.

Figure 3 shows the microprobe analysis of Al, Ce

and Si for the ceria-coated silica-alumina fiber, heated in (a) an Ar atmosphere and (b) subsequently in air at 1100°C. The Al concentration along the electron beam scanning line in the cross section of the fiber Fig. 3(a) was almost constant, indicating a uniform distribution of Al. On the other hand, the Ce concentration in Fig. 3 (a) showed a peak in the ceria layer coating on the surface of the fiber. In addition, the Ce concentration was not flat along the scanning line in the cross section and became lowest at the center of the fiber. This observation indicates that the Ce ions diffused from the surface to inside of the fiber. The Si concentration along the line became higher in the center and in the ceria layer on the fiber, indicating the diffusion of Si<sup>4+</sup> ions from inside to the surface to form 7Ce<sub>2</sub>O<sub>3</sub>·9SiO<sub>2</sub>.



Fig. 3. Microprobe analysis of Al, Ce and Si for the ceria-coated silica-alumina fiber, heated in (a) an Ar atmosphere and (b) subsequently heated in air at 1100°C.



Fig. 4. Microprobe analysis of Si, Al, Ce and Yb for Yb-doped ceria coated SiC fiber, heated in an Ar atmosphere at 1100°C.

The above analysis leads to the following conclusion that (1) CeAlO<sub>3</sub> was formed inside the fiber through the diffusion of Ce<sup>3+</sup> ions from the surface of the fiber (Eq. (3)) and (2)  $7Ce_2O_3 \cdot 9SiO_2$  was formed on the ceria layer of the fiber through the interdiffusion of Si<sup>4+</sup> and Ce<sup>3+</sup> ions. The change of the Al, Si and Ce concentrations after the subsequent heating in air at 1100°C is shown in Fig. 3(b). The decrease of the fiber suggests that Si<sup>4+</sup> and Ce<sup>3+</sup> ions diffused towards the inside and surface of the fiber, respectively, after the decomposition of CeAlO<sub>3</sub> (Eq. (3)) and  $7Ce_2O_3 \cdot 9SiO_2$  (Eq. (4)).

Figure 4 shows the microprobe analysis of Si, Al, Ce and Yb for the Yb-doped ceria-coated SiC fiber, heated in an Ar atmosphere at 1100°C. The Si concentration inside the fiber was almost constant. It was difficult to characterize the Al concentration gradient because of the low content (< 1 mass%). The Ce and Yb concentrations were almost constant inside the fiber and became higher at the coated layer. This observation suggests that the reaction between the SiC fiber and the coated layer occurred only at the interface and the interdiffusion of Ce<sup>3+</sup> and Si<sup>4+</sup> ions was limited to near the interface. For this reason, no significant change of Ce and Yb concentrations was observed in the cross section of the fiber after the subsequent heating in air.

# Microstructures and mechanical properties of SiC fibers coated with Yb-doped ceria/mullite-alumina matrix composite

The ball-milled mullite-alumina powder with a cumulative particle size distribution of 0.5 µm/10%-1.2 µm/ 50%-2.4 µm/90%, was formed into sheets of 75 µm thickness by a doctor blade method. The unidirectional SiC fibers coated with Yb-doped ceria were distributed on the mullite-alumina sheets. The laminated green composite with 15.2 vol% SiC fibers was densified to a relative density of 90.7% by hot-pressing at 1200°C for 2 h in an Ar atmosphere. Figure 5 shows typical stressstrain curves for the hot-pressed composite (a) and for the composite subsequently heat-treated at 1100°C for 2 h in air (b). As seen in Fig. 5, the deformation behavior of the composite depended on the processing atmosphere. In the case of hot-pressing in an Ar atmosphere, the composite showed nonlinear stress-strain curves with low Young's modulus of 174 GPa and fractured at a stress of 90 MPa and at a strain of 0.08 %. The subsequent heat-treatment in air provided a brittle composite with a high Young's modulus of 353 GPa and flexural strength of 187 MPa. The above difference in the mechanical properties is related to the phase stability of the interfacial material (Yb-doped ceria). As seen in Fig. 2, CeAlO<sub>3</sub> and 7Ce<sub>2</sub>O<sub>3</sub>·9SiO<sub>2</sub> were formed by the chemical reaction between the Ybdoped ceria and SiC fibers in an Ar atmosphere. The



**Fig. 5.** Typical stress-strain curves for the hot-pressed composite (a) and for the composite subsequently heat-treated at 1100°C for 2 h in air (b).

formation of CeAlO<sub>3</sub> is also expected between the Ybdoped ceria and alumina in the matrix. This type of chemical reaction forming an interfacial compound usually strengthens the interfacial bonding between the fibers and matrix, leading to brittle fracture of the composites. However, the SiC fibers/Yb-doped ceria/ mullite-alumina system heated in an Ar atmosphere produced a ductile composite. The possible reasons are (1) the formation of a mechanically weak interfacial compound, and (2) the formation of a porous interface layer. The high porosity (~60%) of the Yb-doped ceria layer may be an important factor in the ductility. The volume change of the solid phase from Eq. (7) in an Ar atmosphere was calculated to be 104%. That is, the solid volume near the interfaces expands slightly with the chemical reaction. In addition, a small chemical interaction is expected between the SiC fibers and the newly-formed solid such as 7Ce<sub>2</sub>O<sub>3</sub>·9SiO<sub>2</sub> or CeAlO<sub>3</sub>. The above factors enhance the ductile properties of the composite. The change to a brittle composite after the heat-treatment in air was accompanied by the disappearance of CeAlO3 and 7Ce2O3.9SiO2 and crystallization of SiO<sub>2</sub> and CeO<sub>2</sub>. The SiC fibers were also oxidized to SiO<sub>2</sub> during the heat-treatment in air. The oxidation of interfacial compounds followings Eqs. (3) and (4) and oxidation of SiC causes a volume expansion near the interfaces (volume expansion: 113% for Eq. (3), 109% for Eq. (4)). Furthermore, the large chemical interaction between the SiO<sub>2</sub> formed and SiC fibers oxidized or mullite-alumina matrix results in strong contact, explaining the increase of Young's modulus by oxidation. No crystallization of alumina in the composites heated in air at 1100°-1300°C suggests the formation of an amorphous SiO<sub>2</sub>-Al<sub>2</sub>O<sub>3</sub> phase,



Fig. 6. Fractured surface of the composite hot-pressed in an Ar atmosphere (a) and subsequently heated in air at  $1100^{\circ}$ C (b).

which also increases the interfacial bonding. The above factors suppress the sliding effect of SiC fibers.

Figure 6(a) shows the fractured surface of a composite hot-pressed in an Ar atmosphere. The pull out of fibers was observed in the fiber yarn, reflecting the weak interfaces. In addition, no crack was produced in the mullite-alumina matrix, suggesting that small residual stresses were induced at room temperature because of the similar thermal expansion coefficients between the fibers and matrix. A similar fractured surface was observed in the composite subsequently heated in air at 1100°C (Fig. 6(b)). Although a slight pull out of the fibers was observed, the composite showed a brittle fracture because of the increase of interfacial bonding.

#### Conclusions

Chemical reactions occurred between the SiC fibers containing a small amount of Al (< 1 mass%) and Ybdoped ceria in an Ar atmosphere at 1100-1300°C, producing CeAlO<sub>3</sub> and 7Ce<sub>2</sub>O<sub>3</sub>·9SiO<sub>2</sub>. The subsequent heating of the SiC fibers in air at 1100°-1300°C decomposed CeAlO<sub>3</sub> and 7Ce<sub>2</sub>O<sub>3</sub>·9SiO<sub>2</sub> into CeO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub> and SiO<sub>2</sub>. The SiC fibers with Yb-doped ceria were distributed on mullite-alumina sheets of 75  $\mu$ m thickness. The laminated composite with 15.2 vol% SiC fibers was densified to a relative density of 90.7% by hot-pressing at 1200°C in an Ar atmosphere. The composite showed ductile properties under a four-point flexural strength test. Pull out of the fibers was observed on a fractured surface. The subsequent heat-treatment of the composite in air at 1100°C resulted in brittle fracture. The dependence of mechanical properties of the composite on the processing atmosphere was correlated to the chemical stability of the interfacial material (Yb-doped ceria).

### References

- R. J. Kerans, R. S. Hay and N. J. Pagano, Am. Ceram. Soc. Bull. 68[2] (1989) 429-449.
- R. J. Kerans and T. A. Parthasarathy, Composites Part A, 30 (1999) 521-524.
- B. Sudre, A. G. Razzell, L. Mollies and M. Holmquist, Ceram. Eng. Proc. 19[4] (1998) 273-280.
- 4. E. D. Morgan and B. Marshall, J. Am. Ceram. Soc. 78[6]

(1995) 1553-1563.

- G. Cain, R. L. Cain and M. H. Lewis, J. Am. Ceram. Soc. 80[7] (1997) 1873-1876.
- M. K. Cinibulk and R. S. Hay, J. Am. Ceram. Soc. 79[5] (1996) 1233-1246.
- R. L. Dong and Y. Hirata, J. Ceram. Soc. Japan, 108[9] (2000) 823-829.
- 8. T. Ishikawa, Y. Kohtoku, K. Kumagawa, T. Yamamura and T. Nagasawa, Nature, 391 (6669) (1998) 773-775.
- T. Ishikawa, S. Kajii, K. Matsunaga, T. Hogami, Y. Kohtoku and T. Nagasawa, Science 282 (1998) 1295-1297.
- Y. Kamino, Y. Hirata and S. Kamata., Proceedings of the 6 th Tohwa University International Symposium, Frontier Nanostructured Ceramics 1996, pp. 165-170.
- Y. Kagawa, "Ceramic Composites", in Composites, Edited by M. Douyama and R. Yamamoto, (Tokyo University, Tokyo, 1984) pp. 165-190
- K. Higashi, K. Sonoda, H. Ono, S. Sameshima and Y. Hirata, J. Mater. Res. 14[3] (1999) 957-967.
- A. C. Tas and M. Akinc, J. Am. Ceram. Soc. 77[11] (1994) 2953-2960.