
EFFECT OF REINFORCEMENTS ON MECHANICAL AND THERMAL PROPERTIES OF SiC SHORT-FIBRE-REINFORCED SiC COMPOSITES

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ABSTRACT

Tyranno SA fibre was higher strength at high temperatures and higher thermal conductivity than that of Hi-Nicalon fibre. Therefore, the aim of this study was to investigate the effect of reinforcements on the mechanical and thermal properties of SiC/SiC composites containing non-coated Tyranno SA short fibres. SiC/SiC composites containing 30 vol.% short fibres were fabricated by tape-casting and hot-pressing at 1650°C-1750°C under a pressure of 40 MPa using an Al_2O_3 - Y_2O_3 -CaO mixture as sintering additives independent of fibre type.

Keywords: SiC/SiC composite, Short fibre, Fracture toughness, Thermal conductivity, Mechanical property

1. INTRODUCTION

Silicon carbide (SiC) fibre has been used as reinforcement in advanced ceramic matrix composites, which are applied in the fields of aerospace, nuclear industry, as high-temperature materials, such as gas-turbine engine parts. This is because SiC fibres exhibit an excellent mechanical behaviour and creep resistance at high temperature with relatively good oxidation resistance. [1]

The process routes of most common continuous-fibre-reinforced composites adopted CVI, PIP or MI. [2-5] They are already studied and partly commercially manufactured for last one decade, but due to their extremely extensive costs, caused by long manufacturing times with special sophisticated techniques, applications are still very restricted up to now. They are only applied in cost-tolerant branches like aerospace applications. Therefore, to open the advantages of fibre-reinforced composites to a broader field of applicants efforts are made to lower the production cost by means of short-fibre-reinforced composites. [6]

In this study, fabrication of SiC short-fibre-reinforced SiC composites using tape-casting and hot-pressing techniques was investigated. Tape casting is a useful process for preparing thin ceramic green sheets. It has been widely adopted to produce ceramic substrate, multi-layered capacities, solid oxide fuel cell, etc. The hot-pressing method is widely used in

commercial production process and it offers the ability to fabricate dense composite. [7] Although the failure of the composite made in this study is still catastrophic, the short-fibre-reinforced composites have generated considerable interest because of the relatively simple manufacturing process, namely, the powder metallurgy method. [8]

The aims of this study were to draw relationships of fibre and matrix in the SiC short-fibre-reinforced SiC composites. Two reinforcement fibres were selected, BN-coated Hi-Nicalon fibre and non-coated Tyranno SA fibre. The effects of processing parameters on mechanical and thermal properties of the SiC/SiC composites using Al_2O_3 - Y_2O_3 -CaO mixture as sintering additives were discussed.

2. EXPERIMENTAL PROCEDURE

The high strength and high modulus SiC fibre used in this study was Hi-Nicalon fibre (Nippon Carbon Co., Japan) and Tyranno SA fibre (Ube Industry Co., Japan), whose chemical composition and typical properties are summarized in Table 1. The starting powder for the matrix components was prepared by ball-milling of a submicron β -SiC powder (Ultrafine, Ibiden, Japan) and various kind of sintering additives with a dispersing agent for 24 h in ethanol using Al_2O_3 balls. The green sheets, about 300-350 μm in thickness, were prepared using a laboratory scale doctor-blade equipment, and stacked along the same direction of the sheet forming. Thus the short fibres

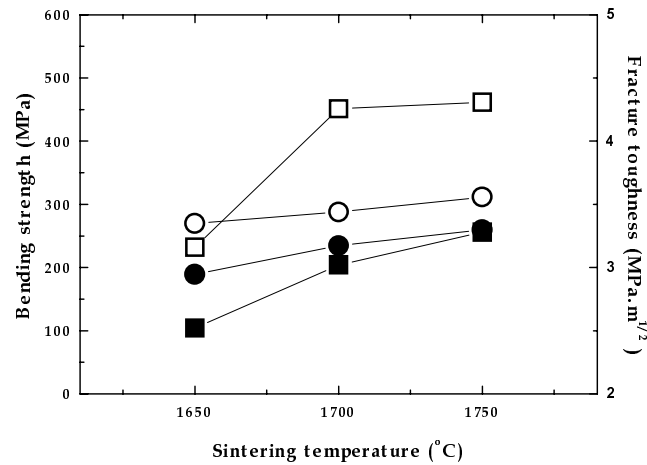


Fig. 1: Maximum strength and fracture toughness of the SiC/SiC composites containing 30 vol.% short fibres sintered at 1750°C. [Open symbol: maximum strength, close symbol: fracture toughness, rectangle: Tyranno SA, circle: Hi-Nicalon]

Table 1: Typical properties and chemical composition of the Hi-Nicalon and Tyranno SA fibres.

Type /SiC fibre	Hi-Nicalon fibre	Tyranno SA
Density /g/cm ³	2.74	3.02
Diameter /μm	14	10
Tensile strength /GPa	2.8	2.8
Young's modulus /GPa	270	420
Elongation /%	1.0	0.7
Specific resistivity /Ohm-cm	1.4	-
Thermal conductivity at R.T.	7.8	64.6
Chemical composition /wt. %	Si: C: O = 63.7 : 35.8 : 0.5	Si: C: O = 67.8 : 31.3 : 0.3

were mostly aligned unidirectionally. The experimental procedures for the fabrication of green sheet have been described elsewhere. [9] The fibre volume fraction of the SiC/SiC composites was about 30 vol.%. The stacked green body was hot-pressed at 1650-1750°C in Ar under a pressure of 40 MPa for 1h.

Bulk density of the specimens was measured by the Archimedes method using water. Three-point bending test (test bars 3.5 x 2.5 x 34 mm) was carried out with a crosshead speed of 0.1 mm/min and a lower span of 30 mm at room in air and high temperatures in vacuum. Bending bars were cut parallel to the fibre axis. Fracture toughness was measured by single-edge-notched-beam (SENB) method with a crosshead speed of 0.05 mm/min at room temperature, and the width of notch was less than 0.2 mm. All data were averages of two or three tests for each condition.

Fracture surface after bending test was observed by scanning electron microscopy (SEM: Hitachi S-3500H). The microstructure observation and composition analysis were carried out in a transmission electron microscopy (TEM: Hitachi H-9000) equipped with an energy-dispersive X-ray spectrometer (EDS: Kevex Delta III).

Hot-pressed specimens were machined into disks (10 mm in diameter and 1 mm in thickness) for thermal conductivity measurement. Thermal diffusivity (α) and specific heat (C_p) were measured by the laser flash method (LF/TCM-FA8510B, RIGAKU, Japan) at room temperature in vacuum (10^{-3} Pa). Thermal conductivity (κ) was calculated from the following equation: $\kappa = \alpha C_p \rho$, where ρ is the bulk density. Thermal conductivity of the composites was measured perpendicular to the direction of fibre axis.

3. RESULTS AND DISCUSSION

Bulk density of the SiC/SiC composites was almost same value regardless of sintering temperature, but density of these composites with Tyranno SA fibre was slightly greater than that of the composites with Hi-Nicalon fibre. This can be attributed to the difference in density of fibres (see Table 1). At 1650°C, these composite achieved nearly full density with Al_2O_3 - Y_2O_3 -CaO as sintering additive. The mechanism of densification of SiC using Al_2O_3 - Y_2O_3 -CaO as sintering additives has reported as liquid-phase-assisted sintering. [10] The low temperature eutectic of the oxides based on Si-Al-Y-Ca-O is an effective densification additive for the fine-grained β -SiC at relatively low temperature. [11]

The bending strength and fracture toughness of the SiC/SiC composites containing 30 vol.% fibres as a function of sintering temperature are shown in Fig. 2. The bending strength of these composites mostly increased with increasing sintering temperature regardless of reinforcements. The bending strength markedly improved after sintering at higher than 1700°C in the case of Tyranno SA fibre reinforced composites. This change cannot be attributed to change in density, which is almost same. It is recognized that composite strength before fracture

was governed by the strength of matrix and strength fibre. Strength of matrix was increased with increasing sintering temperature in this matrix composition from the comparative experiments for monolithic ceramics with same additive composition with present study. [12] The fracture toughness of these composites slightly increased with increasing sintering temperature, which that of the composites hot-pressed at 1750°C was about $3.25 \text{ MPa m}^{1/2}$ regardless of reinforcements. The reason for the lower fracture toughness of Tyranno SA fibre-reinforced composites comparing to Hi-Nicalon fibre-reinforced composites could be explained as follows. Crack propagation from matrix to fibre is not attributed because interface between fibre and matrix was bonded strongly due to reaction with sintering additives. Therefore, fibre pull-out could not be observed as mentioned later.

Fig. 2 shows the microstructures observation by SEM photographs of the SiC/SiC composites sintered at 1750°C. As shown in SEM photograph, in the case of the Hi-Nicalon fibre-reinforced composite, interface of matrix/matrix could be observed as a void without reaction between fibre and matrix. In the case of Tyranno SA fibre-reinforced composite, it could be difficult to make a clear distinction of interface because of reaction of the fibre and oxide components of sintering additives. The reason for the difference of the interface in two composites could be attributed for the existence or not (BN-coated Hi-Nicalon fibre and non coated Tyranno SA fibre) of coating layer of fibre before composite fabrication. Boron nitride (h-BN) has unique chemical and physical properties such as low density, high melting point, chemical inertness, graphite-like layer structure, and easy to peel. Boyen et al. [13] reported that h-BN was stable at high temperature and difficult to react with other components.

Fig. 3 shows the room temperature thermal conductivity of two composites sintered at 1750°C. Thermal conductivity of these composites was too low compared to that of the generally reported SiC ceramics. Thermal conductivity of Tyranno SA reinforced-reinforced composites, about 32 W/m K , was higher than that of Hi-Nicalon reinforced-

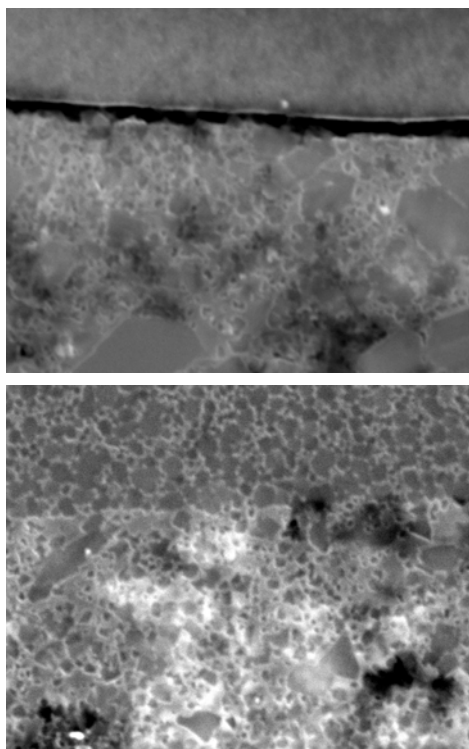


Fig. 2: Microstructures on the matrix/matrix interface of the SiC/SiC composites sintered at 1750°C.

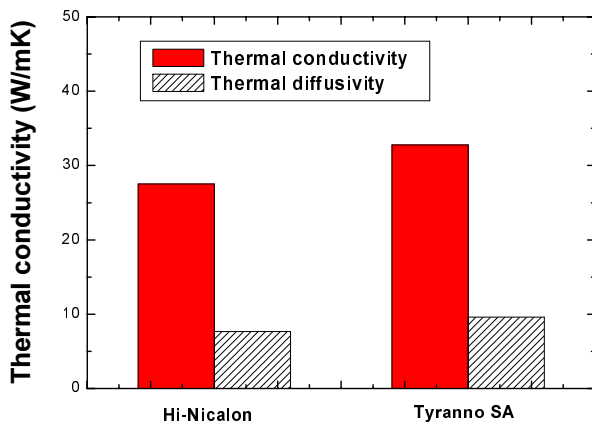


Fig. 3: Thermal conductivity and thermal diffusivity of the SiC/SiC composites sintered at 1750°C.

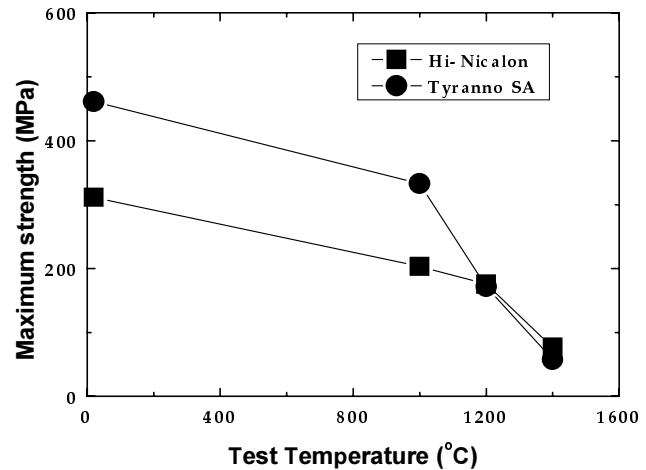


Fig. 4: Maximum strength of the SiC/SiC composites sintered at 1750°C as a function of test temperatures.

reinforced SiC composites. Relatively lower thermal conductivities of Hi-Nicalon fibre-reinforced composite than that of Tyranno SA fibre-reinforced composite can be attributed to lower conductivity of the fibre and BN interface. Thermal conductivity of Hi-Nicalon and Tyranno SA fibre was reported to be 7.8 W/m K, 64.6 W/m K at room temperature, respectively. Further reason for lower conductivity of these composites can be attributed for the large amount of oxide grain boundary phase. [14] Thermal conductivity is important for minimizing the stress in a material exposed to a thermal gradient and for removing heat from energy conversion systems. The thermal conductivity of CMCs is generally less than that of their monolithic ceramics because of matrix/matrix interface and higher porosity. [15] Westwood et al [16] reported that the thermal conductivity of high-quality single crystal SiC 490 W/m K while BeO-doped SiC had a thermal conductivity of 270 W/m K at 25°C and single crystal SiC a conductivity of 90 W/m K at 25-150°C. [17] Thermal conductivity of polycrystalline SiC is a function of variable such as impurity content, processing method, but has been reported to be 12 to 35 W/m K.

The bending strength of these composites sintered at 1750°C as a function of test temperatures is shown in Fig. 4. The bending strength was decreased with increasing test temperature regardless of reinforcements. At higher temperatures, maximum strength was mostly maintained up to 1000°C, but decreased at 1200°C in the all samples. In the case of the Hi-Nicalon fibre-reinforced composite, maximum

strength was about 370 MPa at room temperature and about 88 MPa at 1400°C, whereas in the case of the Tyranno SA fibre-reinforced composite, maximum strength was about 450 MPa at room temperature and about 86 MPa at 1400°C. Maximum strength of the Tyranno SA fibre-reinforced composite was higher than that of the Hi-Nicalon fibre-reinforced composite at room temperature up to 1000°C. Degradation of composite strength above 1200°C can be attributed to the reduction of matrix strength. The fracture behaviour of the SiC/SiC composites showed completely brittle fracture from room and high temperatures regardless of reinforcements. The reason for brittle fracture of these composites could mainly be explained by the degradation of fibre, as mentioned below, and strengthening of matrix/matrix interfacial bonding. Although Hi-Nicalon fibre used in this study was coated with BN, the coating does not work as weak boundary between fibre and matrix. In the case of Tyranno fibre, nothing was coated. In this case, weak bonding was not formed. The other factor for brittle fracture particularly, in the case of Tyranno SA fibre reinforced composite, is strength of matrix. Matrix strength was much higher than that of Hi-Nicalon fibre reinforced composite, therefore, more fibre strength/volume fraction should be necessary. The mechanical strength of SiC fibres should be degraded by the exposure at high temperature. [18] Yoshida et al. [19] reported that fracture behaviour of the continuous-reinforced-reinforced SiC composite fabricated by hot-pressing at 1750°C with 40-52 vol.% fibre showed ductile fracture at room and high temperatures. Maximum

strength of their composites with non-coated and BN-coated fibre was about 240 MPa and 168 MPa, respectively. These results indicated that maximum strength of the short-fibre-reinforced composites with the same sintering additive and sintering temperature was higher than that of the continuous-fibre-reinforced composites, particularly in the case of Tyranno SA fibre, but fracture behaviour of the short-fibre-reinforced composites was catastrophic.

Fracture surfaces of the SiC/SiC composite sintered at 1750°C as a function of reinforcements and tested at room and high temperatures are shown in Fig. 5. It was observed that fibres were mostly aligned along sheet plane and long axis of the bend bars, i.e., perpendicular to the hot-pressing direction. As indicated in the Fig., in the case of Hi-Nicalon fibre-reinforced composite, slight pull-out of short fibres was observed, but fracture behaviour was catastrophic failure. On the contrary, in the case of Tyranno SA fibre-reinforced composite, that of short fibres was not observed. Furthermore, in the case of Tyranno SA fibre-reinforced composite, round fibre

morphology was not clear in the composites sintered at 1750°C, and fracture surface of fibres showed polycrystalline feature. The reason for the difference in fibre pull-out property was attributed to existence of interface between fibre and matrix. The weak interface can induce crack deflection along the interface, permitting intact fibres to bridge crack faces. In the present system, relatively strong bonding between the fibre and the matrix, not only Tyranno fibre-reinforced composites but also BN coated Hi-Nicalon fibre-reinforced composites, is probably formed, therefore, pull-out of fibres are not sufficient. [20]

Fig. 6 shows the TEM micrographs of the matrix/matrix interface of the SiC/SiC composites sintered at 1750°C. As shown in the Fig., BN compound in the interface of the Hi-Nicalon fibre-reinforced composite was observed. From the results of EDS analysis, the present of C, O, Al, Si elements in the

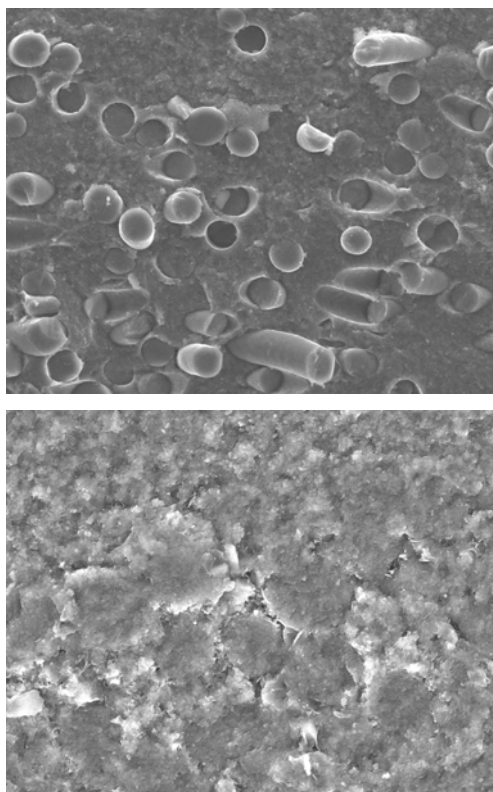


Fig. 5: Fracture surfaces of the SiC/SiC composites sintered at 1750°C. (a) Hi-Nicalon reinforced-reinforced-, (b) Tyranno SA reinforced-reinforced composite, tested at room temperature

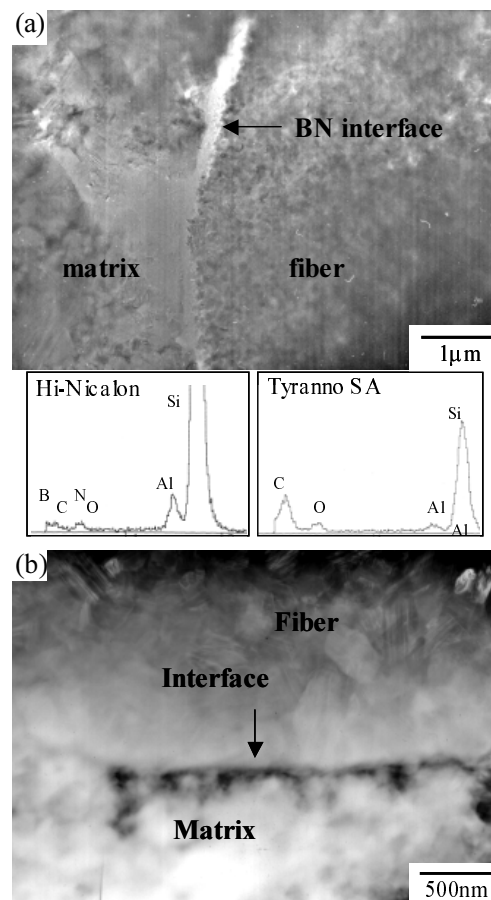


Fig. 6: TEM micrographs on the matrix/matrix interface of the SiC/SiC composites sintered at 1750°C.

interface of Tyranno SA fibre-reinforced composite was detected, whereas that of C, O, B, N, Si elements in the interface was detected in the interface of Hi-Nicalon fibre-reinforced composite, respectively. It is indicated BN interface layer of Hi-Nicalon could not work as a diffusion barrier for sintering aids for matrix densification.

4. CONCLUSIONS

SiC/SiC composites containing 30 vol.% short fibres were fabricated by tape-casting and hot-pressing at 1650°C-1750°C under a pressure of 40 MPa using an Al_2O_3 - Y_2O_3 -CaO mixture as sintering additives independent of fibre type. The composites fabricated by this process achieved nearly full density at 1700-1750°C. The bending strength increased with increasing sintering temperature regardless of reinforcements and decreased with increasing testing temperature. The maximum strength of Tyranno SA fibre-reinforced SiC composites, which was about 460 MPa, was higher than that of Hi-Nicalon fibre-reinforced SiC composites. Fracture toughness was slightly increased with increasing sintering temperature regardless of reinforcements. The fracture toughness of SiC/SiC composites hot-pressed at 1750°C was about $3.3 \text{ MPa m}^{1/2}$. In the case of Hi-Nicalon fibre-reinforced composite, slight pull-out of short fibres was observed, but fracture behaviour was brittle failure. On the contrary, in the case of Tyranno SA fibre-reinforced composite, that of short fibres was not observed. Thermal conductivity, about 32 W/m K, of Tyranno SA fibre-reinforced composites was higher than that of Hi-Nicalon fibre-reinforced SiC composites. Relatively lower thermal conductivities of Hi-Nicalon fibre-reinforced composite than that of Tyranno SA fibre-reinforced composite can be attributed to lower conductivity of the fibre and BN interface.

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