JOURNAL OF MATERIALS SCIENCE **34** (1999) 1505– 1511

Mechanical properties of Si-Ti-C-O fibre-bonded ceramic using satin weave

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Tensile and compressive properties of fibre-bonded ceramic (Tyrannohex) are reported along with the relationship between fibre orientation and flexural strength. In this study, Satin-Tyrannohex was produced by hot-pressing an oxidized satin-woven Tyranno fibre. The Satin-Tyrannohex showed the most well-balanced mechanical properties in all directions from the fibre axis at room temperature compared with crossplied- or 0°/±45°/90°-Tyrannohex. The Satin-Tyrannohex maintained excellent strength up to 1400 °C, which is comparable to or greater than the strength at room temperature. Decreases in tensile and compressive strengths at 1500 °C could be due to an increase in the critical length of the fracture fibre and a decrease in the capability to support the fibre, respectively, because of the slight "softening" of the matrix. © *1999 Kluwer Academic Publishers*

1. Introduction

Development of the thermostructural materials, which withstand long-term use in corrosive environments at high temperatures, has been expected by aerospace industries, who require greater energy efficiency and additional functionality [1–3]. Ceramic matrix composites (CMCs) reinforced with continuous fibres have been identified as the most promising candidate of these thermostructural materials, because they guarantee a high heat-resistance and an excellent fracture toughness. One of such CMCs is Si-Ti-C-O fibre-bonded ceramic (Tyrannohex®) which is produced by hot-pressing sheets or fabrics of a pre-oxidized Si-Ti-C-O fibre with the oxide layer on the surface [4]. Tyrannohex possesses a close-packed structure of raw fibre ($V_f \cong 85$ vol %), with the interstices between the fibres perfectly filled with both the oxide material which initially covered the raw fibres, and very fine TiC particles (<50 nm) uniformity dispersed in this oxide material [5]. Moreover a thin interfacial carbon layer (thickness: ∼15 nm) which is turbostratic in structure exists around all the foregoing fibres.

In the previous research we have investigated the microstructure and mechanical properties of Tyrannohex [6, 7]. Representative results of this research are as follows. The turbostratic interfacial carbon layer formed during the hot-pressing process serves as a slip layer to prevent propagation of cracks generated as a result of the initial fracture of the matrix or the fibres to the intact fibres. In addition, unidirectional Tyrannohex (UD-Tyrannohex) in which all the fibres were aligned unidirectionally along the stress axis showed elastic behaviour nearly to the maximum tensile load. This behaviour occurs because of high volume fraction of fibres, which greatly decreases the effects of the matrix. However, unidirectional composites may not be particularly useful in the case of an actual component, since either proper orientation of fibres along the major stress axis or isotropy in all directions are required. Thus, the evaluation of mainly two-directional Tyrannohex using the fabrics of the raw fibres and to clarify the fundamental behaviour are imperative for future designs.

The purpose of the present study was to clarify the relation among fibre orientation and mechanical properties and performance of mainly two-directional Tyrannohex (Satin-Tyrannohex) fabricated from satin weaves of Si-Ti-C-O fibre. Mechanical properties and performance were evaluated in tension, compression and flexural test at temperatures up to 1500 °C, with fracture mechanisms investigated at each temperature.

2. Experimental procedure

2.1. Material

The reinforcing material used in this study was eight-harness satin weave fabric of Si-Ti-C-O fibres (Tyranno fibre®, LoxM grade, Ube Industries LTD.) containing oxygen of 12 mass%. Before fabrication of the preform, the single Tyranno fibre [8] bundles were oxided in ambient air at 1000 °C for 20 hours. Satin weave fabrics of the pre-oxidized Tyranno fibres which were covered with oxide layer on the surface were prepared. A preform was then prepared by laminating the pre-oxidized satin weaves of Tyranno fibre. Finally,

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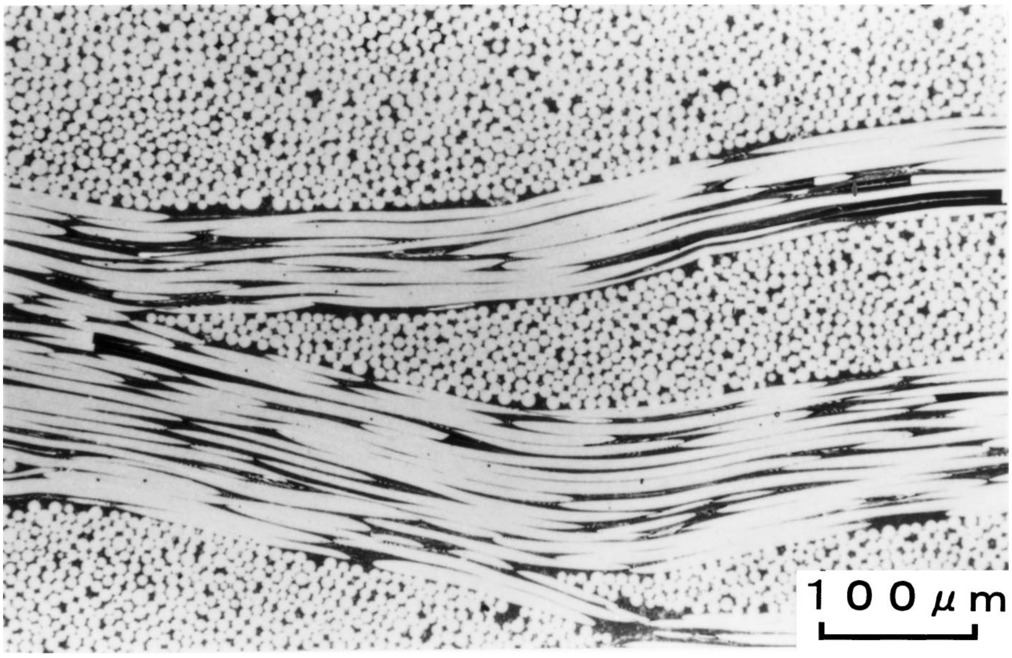

Figure 1 Optical micrograph of a cross section of Satin-Tyrannohex by hot-pressed in argon atmosphere at 1750 °C under 40 MPa.

the Satin-Tyrannohex composite was produced by hot-pressing the preform in argon atmosphere at 1750 °C under 40 MPa of pressure for 1 hour. The final fibre volume fraction (V_f), the density and the porosity are shown in Table I. The very little porosity was confirmed by scanning electron micrography (see in Fig. 1).

2.2. Mechanical testing

The influence of the fibre orientation on the four point flexural strength was examined using test specimens with dimensions of about 4 mm × 3 mm × 40 mm and loading and supporting spans of 10 mm and 30 mm, respectively. Testing was carried out at a crosshead speed of 0.5 mm min^{-1} at room temperature according to JIS R 1601 using a Shimazu Auto-Graph (DSS-500). The strength was calculated from the maximum load.

UD-Tyrannohex, cross-plied Tyrannohex (CP-Tyrannohex) in which fibres were aligned in orthogonal direction every alternate layer using a thin unidirectional sheet, and pseudo-isotropic Tyrannohex (0°/±45°/90°-Tyrannohex) in which fibres were rotated in 0°/±45°/90° directions using unidirectional sheets, were also examined in a comparative point of view.

Tensile tests were performed in air at temperatures up to 1500 °C using the test specimens shown in Fig. 2a. Testing was carried out at a crosshead speed of 0.5 mm min^{-1} after holding at a designated temperature for 15 min. In this test, a universal testing machine (MTS 808) with a hydraulically-operated grip was used. Moreover, the fracture process in tensile test was also observed using another type of test specimen shown in Fig. 2b according to the following procedure. The parallel sides of the test specimen were polished using fine diamond pastes (<1 μm). Pre-cracked specimens loaded up to 135 MPa at room temperature or 1400 °C were prepared. Being prepared at 1400 °C, the specimen was loaded in argon atmosphere. The differences in the crack formation among these specimens were observed using a field-emission scanning electron microscope[†].

Compressive test was performed using the specimen with dimensions of 3 mm × 3 mm × 3 mm in argon atmosphere at the same temperature as the flexural tests. Testing was carried out at a crosshead speed of 0.5 mm min^{-1} after holding at a designated temperature for 15 min. In this test, an universal testing machine (Instron 8562) was used. The compressive strength was examined in the direction in plane. The morphology of the post-fracture surfaces of compressive specimen was examined using a scanning electron microscope[‡].

TABLE I Characterization of Tyrannohex

Type	V_f (%)	Density (g cm^{-3})	Porosity (%)
UD-Tyrannohex	88–90	2.52	<1
Cross plied-Tyrannohex	85–87	2.46	<1
0/±45/90-Tyrannohex	85–87	2.46	<1
Satin-Tyrannohex	85–87	2.46	<1

3. Result and discussion

3.1. Relation between the fibre orientation angle and flexural strength

Relation between the fibre orientation angle and flexural strength of Satin-Tyrannohex is shown in Fig. 3 compared with the results of UD-, CP-, and 0°/±45°/90°-Tyrannohex. Both Satin-Tyrannohex and CP-Tyrannohex have two directional orientation (0°/90°) of the fibre. However, for the strength by flexural testing with longitudinal axis of 45° direction to the fibre direction, Satin-Tyrannohex was greater than that of CP-Tyrannohex. Moreover, of the three

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[†] Model S-4200, Hitachi, Tokyo, Japan.

[‡] Model JST-T220, Jeol-Technics Co., Ltd., Tokyo, Japan.

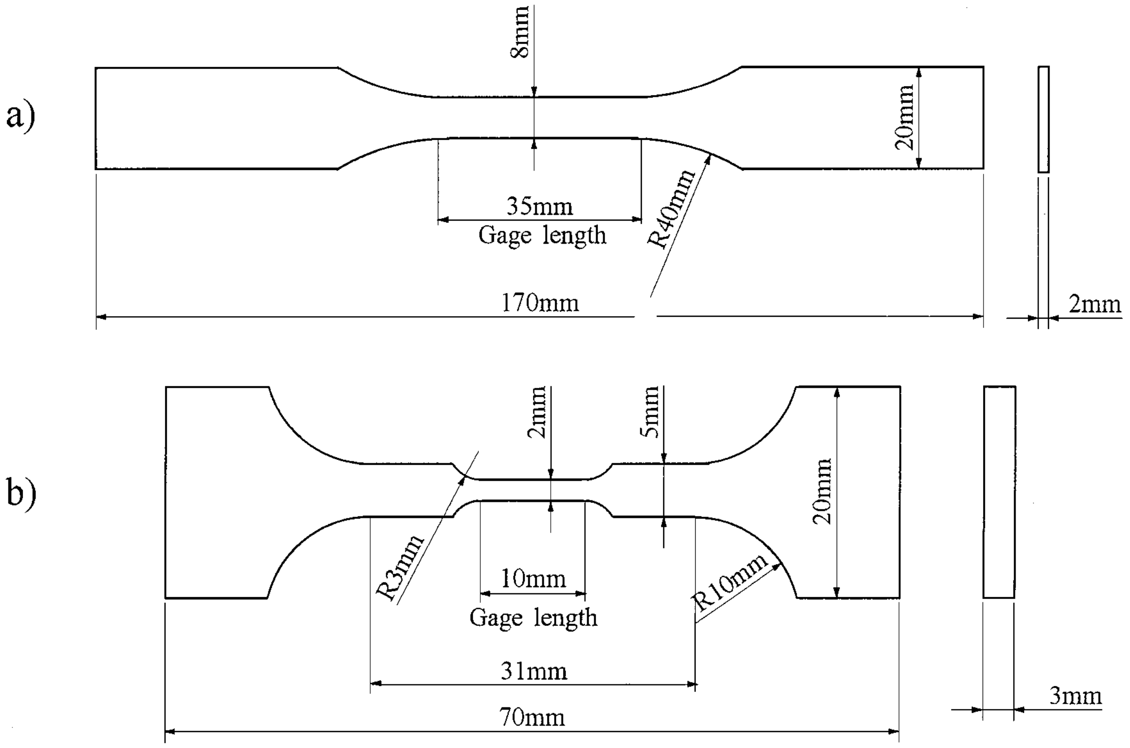

Figure 2 Representative shapes and dimensions of tensile test specimens for (a) a measurement of the temperature-dependence and (b) an observation of the fracture process.

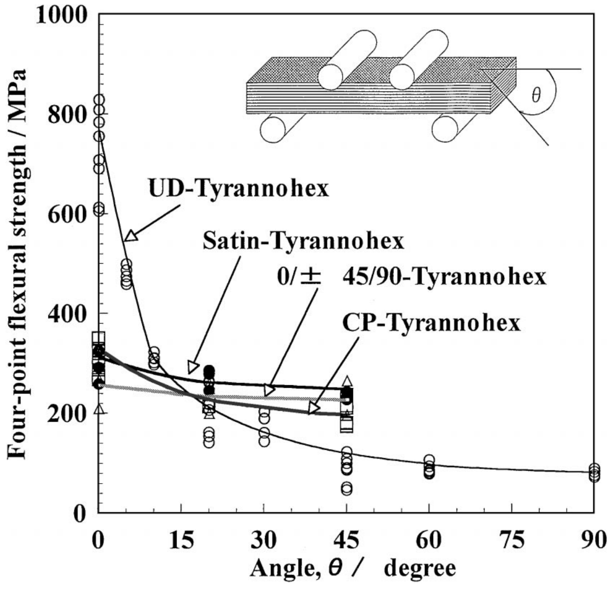

Figure 3 Relation between the fibre orientation angle and flexural strength of Satin-Tyrannohex at room temperature in comparison with that of UD-Tyrannohex, CP-Tyrannohex and 0°/±45°/90°-Tyrannohex.

types of Tyrannohex, Satin-Tyrannohex showed greater strength in all longitudinal test directions compared with 0°/±45°/90°-Tyrannohex. Namely, of all types of Tyrannohex, Satin-Tyrannohex showed the most well-balanced mechanical properties in all directions from the fibre axis at room temperature, because the satin weave has many intertwined fibres in itself. In addition, the woven cloth is very favourable for handling during the preparation process which is extremely advantageous in making Satin-Tyrannohex a practical material.

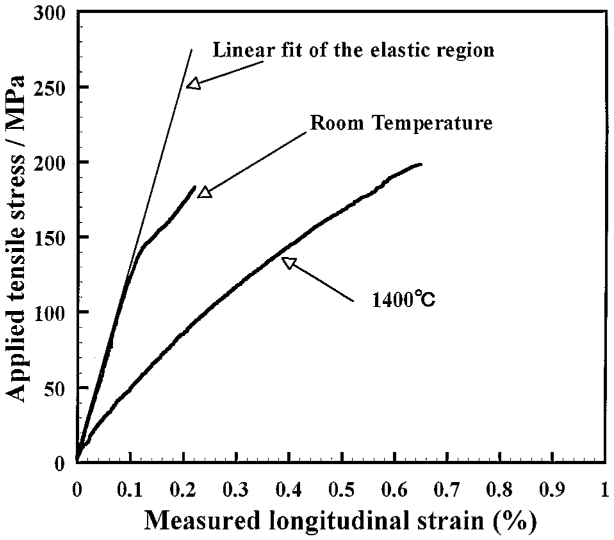

Figure 4 Tensile stress-strain curves of Satin-Tyrannohex at room temperature and 1400 °C.

3.2. Process of tensile fracture at room temperature

Typical stress-strain curves of Satin-Tyrannohex at room temperature and 1400 °C are shown in Fig. 4. At room temperature an abrupt increase in compliance was observed at stresses greater than 130 MPa, whereas the compliance at 1400 °C gradually increased with no abrupt changes.

In order to clarify the reason for the changes in the compliance at each temperature, each fracture pattern was examined. An SEM micrograph of the surface of a tensile specimen at room temperature, for which loading had been stopped at 135 MPa, is shown in Fig. 5. Most of the cracks are shown between the fibres

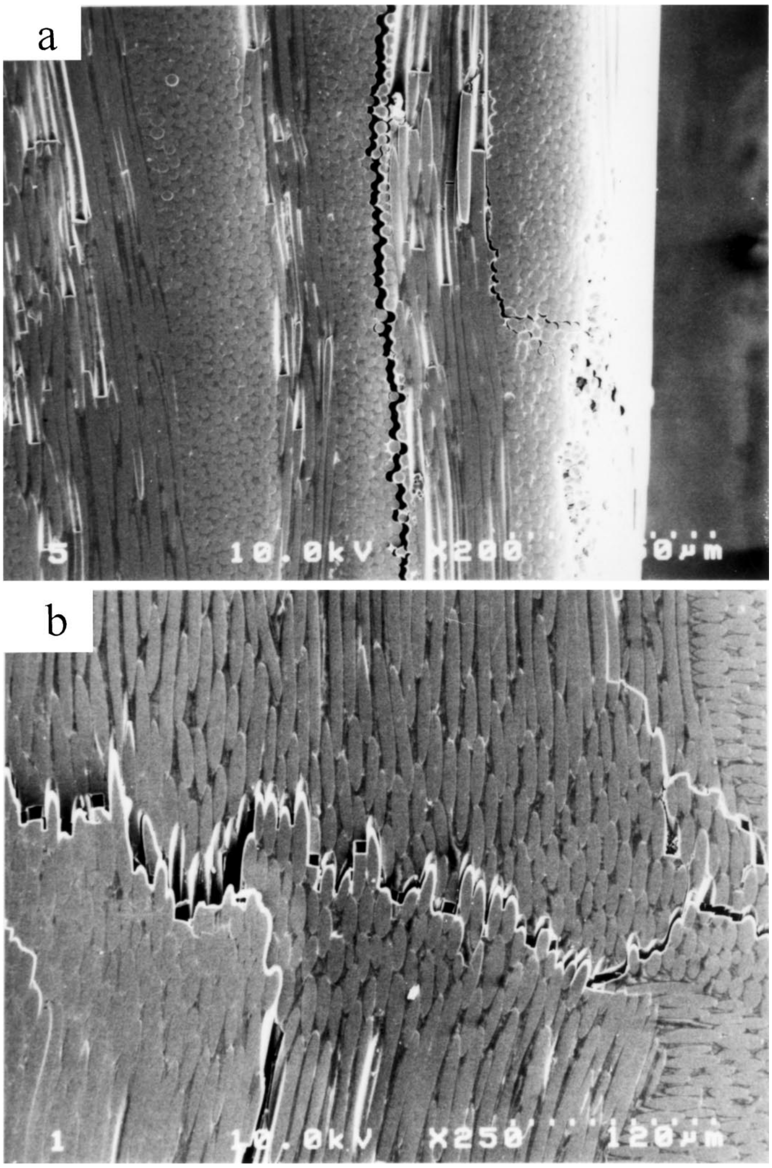

Figure 5 SEM micrographs showing polished sections of Satin-Tyrannohex after loading at the stress of 135 MPa at room temperature; (a) a cross-section perpendicular to the laminating plane; (b) a parallel section to the laminating plane.

transverse to the stress direction (see in Fig. 5a). The majority of these cracks propagated along the interface between the matrix and the fibre, though some cracks propagated in the matrix. The cracks deflected primarily in the vicinity of the surface of the longitudinal fibres. Moreover, a separation of the surface layer of the specimen and a breaking of the longitudinal fibre also occurred (see in Fig. 5b). Thus, on the fracture in the interstices between the fibres transverse to the stress direction, a separation of the surface layer of the specimen and a local breaking of the longitudinal fibres are assumed to be the main cause of the increase in the compliance at room temperature. Likewise, SEM photographs of the surface of the specimen tested in argon atmosphere at 1400 °C, for which the loading had been stopped when the tensile stress reached 135 MPa, is shown in Fig. 6. Cracks are shown in the interstices between the fibres transverse to the stress direction (see in Fig. 6a). These cracks propagated along the interface between the fibre and the matrix and deflected primarily in the vicinity of the surface of the longitudinal fibre. Moreover, breakage of the longitudinal fibre was observed (see in Fig. 6b). Thus, the continuous increase in compliance at 1400 °C is assumed to be due to the cracking in the interstices between the transverse fibres and the local breaking of the longitudinal fibres.

Additional phenomena concerning the abrupt change in compliance at room temperature and the gradually increase in compliance at 1400 °C are going to be discussed as follows whereas difference between the fracture processes at room temperature and 1400 °C are examined in following subsection. Separations of the surface layer of the specimen and fracture of the matrix were not observed at 1400 °C, although they were observed in the specimen tested at room temperature. Thus, these two phenomena are presumed to be the main cause of the abrupt increase in the compliance at room temperature.

Incidentally, the proportional limit of Satin-Tyrannohex (∼130 MPa) was greater than that of the other bi-directional CMCs reinforced with continuous fibres. Although not fully substantiated, this may be due to the following phenomena:

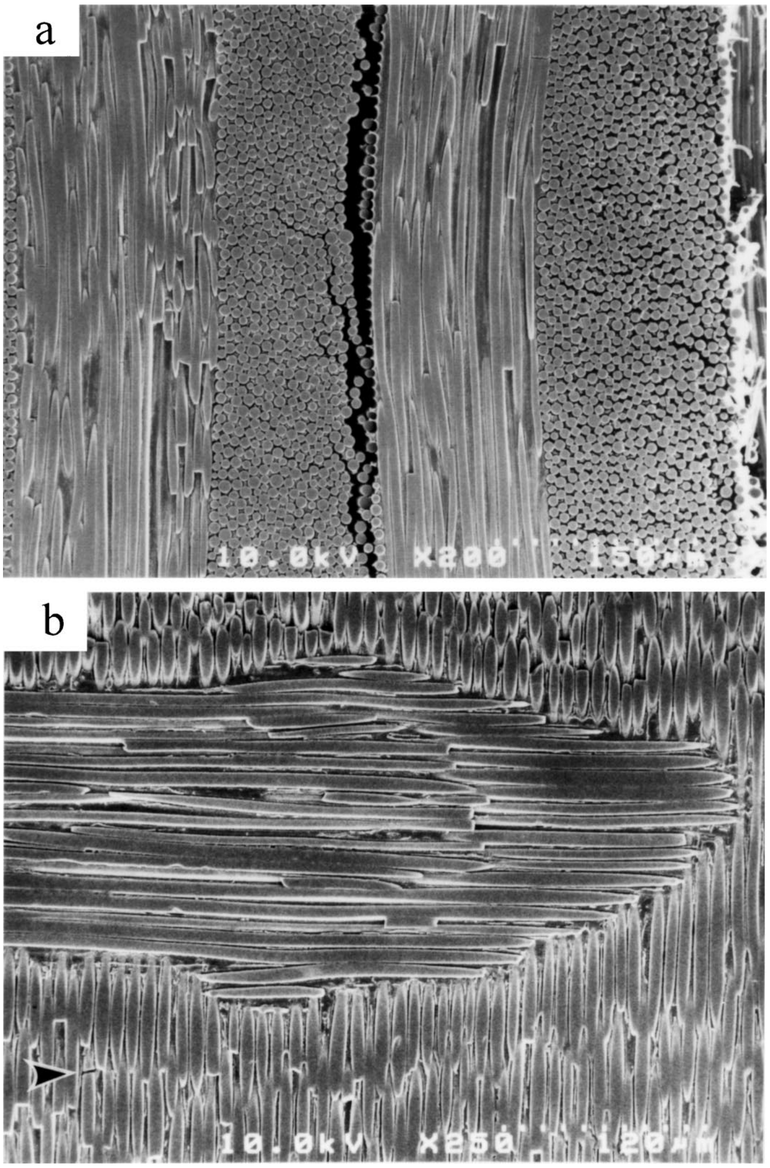

Figure 6 SEM micrographs showing polished sections of Satin-Tyrannohex after loading at the stress of 135 MPa at 1400 °C; (a) a cross-section perpendicular to the laminating plane; (b) a parallel section to the laminating plane.

1. The structure of Satin-Tyrannohex is extremely dense, and there are few defects which may leads the crack formation.
2. The volume of the matrix in Satin-Tyrannohex is extremely low. (Volume fraction of the matrix: $<15\%$)
3. The inter-laminar shear strength (ILSS) of Tyrannohex is relatively high compared with the other CMCs. (UD-Tyrannohex, ILSS ≈ 80 MPa)

3.3. Tensile and compressive strength

Temperature-dependence of the tensile and compressive strength of Satin-Tyrannohex is shown in Fig. 7. The tensile strength as a function of temperature was constant up to 1400 °C, and then only slightly decreased at higher temperatures. The compressive strength as a function of temperature increased slightly up to 1200 and 1400 °C, but then decreased at higher temperatures.

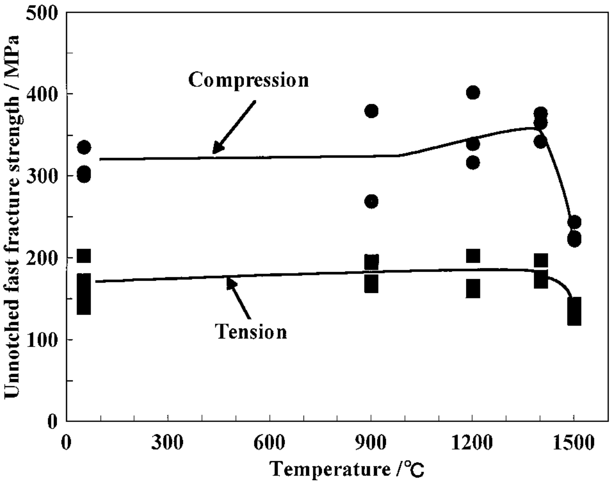

Figure 7 Temperature-dependence of tensile and compressive strength of Satin-Tyrannohex.

In order to clarify the temperature-dependence of the tensile and compressive strength, morphological changes were investigated. To begin with, the reason why the tensile strength decreased at 1500 °C is discussed. Typical fracture morphologies of the specimens tested at room temperature, 1400 and 1500 °C are shown in Fig. 8. Fibre pullout was observed at the

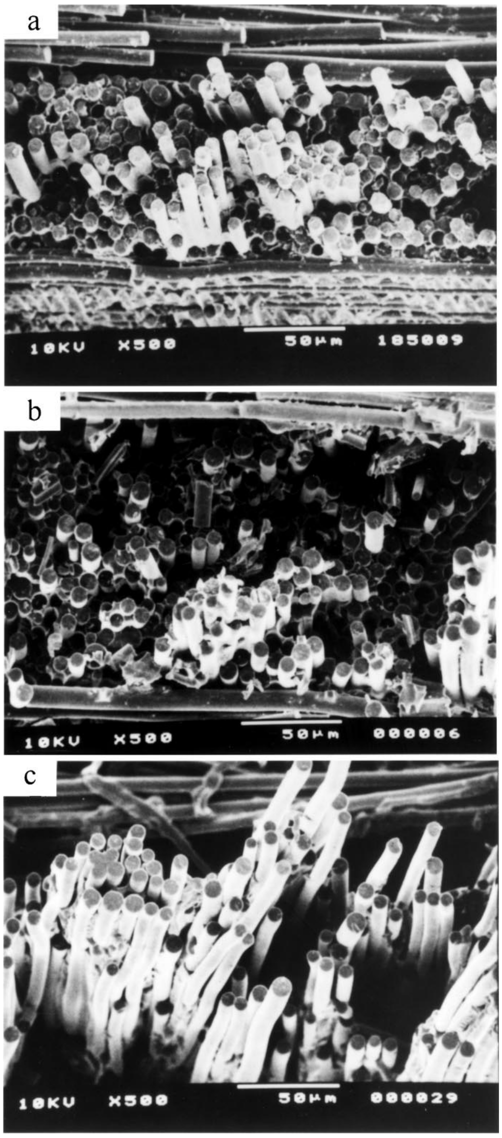

Figure 8 SEM micrographs showing tensile fracture morphology of Satin-Tyrannohex tested at (a) room temperature, (b) 1400 °C and (c) 1500 °C.

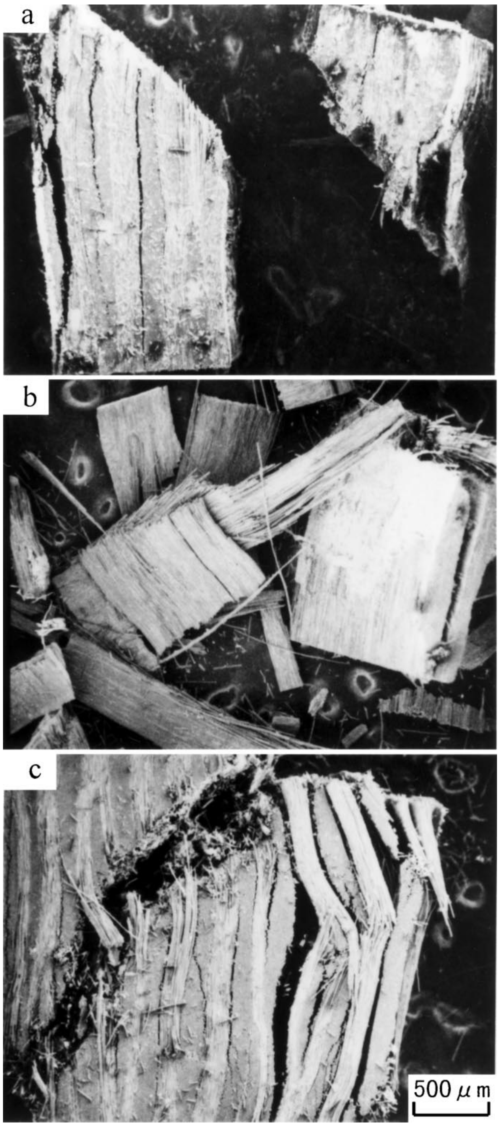

Figure 9 SEM micrographs showing compressive fracture morphology of Satin-Tyrannohex tested at (a) room temperature, (b) 1400 °C and (c) 1500 °C.

fracture surface in each specimen. However, the length of the fibre pullout at 1500 °C was longer than that at 1400 °C. Note that the length of the fibre pullout is closely related with the "adhesive" strength between the fibre and its interface[§]. It is generally known [9, 10] that an increase in the fibre pullout length means a decrease in bond strength between the fibre and the matrix. From a viewpoint of local stress concentration, a decrease in the bond strength causes a decrease in the local stress concentrations, and consequently results in an "increase" in the material strength. On the other hand, from a viewpoint of the reinforcing efficiency, a decrease in the bond strength causes an increase in the critical length of the fracture fibre, and consequently results in a decrease in material strength. In the actual experimental results, the change in the tensile strength corresponded to the change in the length of the fibre pullout. Namely, the length of the fibre pullout increased with a decrease in the tensile strength. In the case of Satin-Tyrannohex, an increase in the critical length of the fracture fibre rather than a decrease in the stress concentration greatly affected its mechanical strength. Incidentally, the decrease in the bond strength between the fibre and matrix is assumed to be due to the "softening" of the matrix. The principal constituent of the matrix is SiO_2 whose strain point[¶] is about 1200 °C [11]. Thus, the decrease in the bond strength at 1500 °C is assumed to be mainly due to a decrease in the shear strength of the matrix rather than a decrease in the adhesive strength between the fibre and the matrix.

Next, in order to clarify the reason for temperature-dependence of the compressive strength, each fracture pattern is discussed. Typical fracture morphologies of the specimens tested at room temperature, 1400 °C, and 1500 °C are shown in Fig. 9. A compressive fracture

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[§] In this paper, the all region between the fibres or the fibre and the matrix is discussed in a lump, though, strictly speaking, they should be divided into the bonding strength between the fibre and the carbon layer, the shear strength of the carbon layer, and the bonding strength between the carbon layer and the matrix on the occasion of discussion about the interfacial strength.

[¶] Strain point is defined as a temperature at which the strain speed reaches 0.0043 mm $\min^{-1}$ [11].

of the fibre along shearing direction to the compression axis and a delamination between laminated weaves were observed at room temperature (see in Fig. 9a). At 1400 °C, crack propagation along the fibres in the longitudinal direction and a burst of the composite into small fragments were observed. A buckling of fibres in the longitudinal direction which supported the compressive load was observed at 1500 °C in addition to compressive fracture along the shearing direction to the compression axis and the delamination between laminated weaves. As aforesaid, the strain point of SiO_2 which is the main component of the matrix is about 1200 °C, thus, an increase in the compressive strength at 1400 °C could be due to an inhibition of the crack propagation by a slightly softened matrix. On the other hand, a decrease in its strength at 1500 °C could be due to a decrease in the capability of support fibres rather than the effect of an inhibition of the crack propagation by more softened matrix.

Incidentally, this difference in behaviour was shown in temperature-dependence of the tensile and the compressive strengths. It is not clear whether the tensile strength increased at about 1200 °C and also 1400 °C, although the compressive strength increased at these temperatures. The decrease in tensile strengths at 1500 °C were small compared with those in the compression strengths. Accordingly, it is assumed that changes in the tensile strength at 1200 and 1400 °C could not be detected because of the small degree of temperature-dependence.

4. Summary and conclusion

The mechanical properties of Satin-Tyrannohex using the satin weave were examined in comparison with those of CP- and 0°/±45°/90°-Tyrannohex. The results are summarised as follows.

1. Satin-Tyrannohex showed the most well-balanced mechanical properties in all direction from the fibre axis at room temperature.
2. The separation of the surface layer of the specimen and the fracture of the matrix are assumed to be the main cause of the abrupt change in the compliance of stress strain curves at room temperature. For the gradual increase in compliance of stress strain curves at 1400 °C fracture in the interstices between the transverse fibres and the local breaking of the longitudinal fibres are assumed to be the main cause.
3. Satin-Tyrannohex maintained excellent, steady strength up to 1400 °C, which is comparable to or greater than the strength at room temperature.
4. An increase in compressive strength at 1400 °C could be due to an inhibition of the crack propagation by a slightly softened matrix.
5. Decreases in tensile and compressive strengths at 1500 °C could be due to an increase in the critical length of the fibre and a decrease in the capability to support fibres, respectively.

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Received 23 January
and accepted 4 November 1998