Textures and Microstructures, 1991, Vols 14–18, pp. 389–395
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ORIENTATION ANISOTROPY IN SiC MATRIX OF UNIDIRECTIONAL SiC/SiC COMPOSITE

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1. INTRODUCTION

SiC/SiC ceramic composites are new materials constituted of a SiC matrix reinforced by SiC-based fibres. They are developed mostly for thermostructural applications, given their low density, high mechanical strength and rigidity, and their chemical inertness.

Studies have been recently published concerning the modelling of the thermomechanical behaviour of these materials[1,2]. These works, in the calculation of elastic behaviour and of residual thermal stresses, assume isotropic properties for the SiC matrix. The primary goal of the study presented here was to assess the validity of this assumption. We therefore present the results of a preliminary investigation concerning the determination of preferential orientations in the SiC matrix of unidirectional SiC/SiC composites.

2. MATERIAL

The ceramic composites studied are constituted of a SiC-based Nicalon (NLM 202) fibre architecture embedded in a SiC matrix obtained by chemical vapour infiltration ("CVI"). The samples processed and provided by S.E.P. ("Société Européenne de Propulsion") are parallelepipedic coupons ($10 \times 10 \times 2$ mm^3) with the square face parallel to the fibre axis. It is to be noted that the fibres, which have a diameter of about 14 μm, contain a fairly large amount of oxygen (typically 11 wt.%).

The SiC matrix is deposited by chemical vapour infiltration. This process, which is based on cracking a precursor (methyltrichlorosilane) in contact with the porosities within the fibrous architecture, is carried out at around 1000°C and under reduced pressure. The densification of the composite requires several infiltration cycles, as evidenced by the concentric rings visible on figure 1.

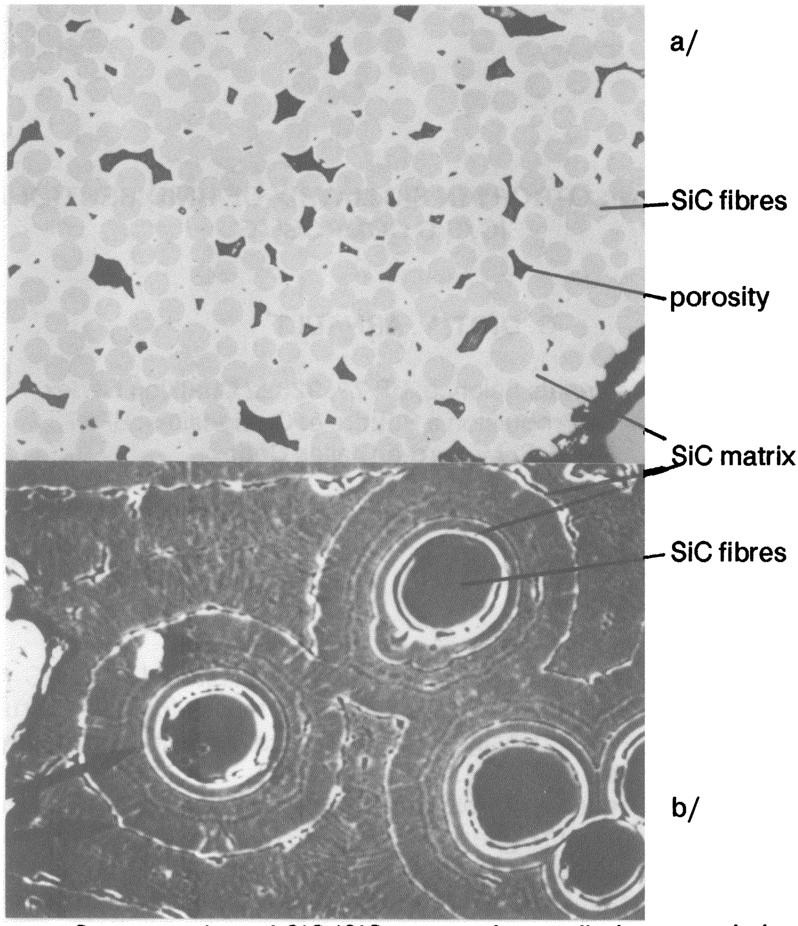

Figure 1. Cross section of SiC/SiC composite studied : general view (a) and detail (b) showing the concentric rings corresponding to successive infiltration cycles (Murakami etchant).

3. EXPERIMENTAL PROCEDURE AND RESULTS

Preliminary remarks

The texture of the SiC matrix has been studied with X-ray diffraction methods. The fact that fibres and matrix are both silicon carbide-based could have rendered difficult or even might have prevented any X-ray investigation on only one of these constituents. Fortunately, as shown by EXAFS studies performed by Laffon et al.[3], the crystallographic domains in Nicalon fibers are very small, with sizes of about 1,5 nm. They do not therefore significantly contribute to X-ray diffraction and the {hkl} peaks in a diffraction diagram of these SiC/SiC composites can be attributed without any ambiguity to the SiC matrix. This makes possible the determination of the matrix texture, as well as its residual stresses.
Moreover it has been verified that the grain sizes in the matrix are fine enough (ranging between 0.1 and a few micrometers) for an adequate statistical study of the grain orientations.

Goniometer θ - 2θ

For this first investigation concerning the determination of preferential orientations in the matrix, the X-ray equipment is an omega-type horizontal Philips PW1380 goniometer equipped with a graphite monochromator. The signals are treated with a PW 1710 electronic device and sent to a Digital PC325 computer.

It is easy with such a goniometer to examine the large faces of a composite sample and therefore to identify the direction normal to these faces. Samples having external surfaces parallel and perpendicular to the fibre axis have been examined. In the latter case, composite specimens had to be specially prepared by stacking together carefully polished strips as indicated schematically on figure 2.

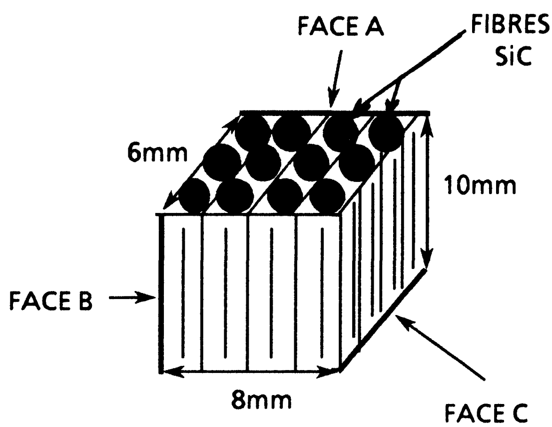

Figure 2 : Composite specimen obtained by stacking unidirectional plates together.

For each of the three selected $\alpha\beta$ directions (perpendicular to faces A, B and C), the largest number of (hkl) reflections is recorded. The corresponding measured intensities cannot, of course, be compared directly.As no isotropic ß-SiC sample was available, the experimental relative intensities have been compared with those listed on 29-1129 ASTM card corresponding to cubic ß-SiC.
The results obtained are presented as relative intensities I/I versus 2θ diagrams on figure 3 for copper K_{α} wavelength (0.15418 nm).

The diagram obtained from face A suggests the presence of a fibre texture : the normals to the {110} and {112} matrix planes are parallel to the SiC fibre axis. In addition to that, the diagrams obtained with faces B and C show that there exists in the SiC matrix a preferred orientation, with {111} planes perpendicular to the SiC fibre axis.

The SiC matrix therefore presents a cylindrical texture that can be schematically represented as on figure 4.

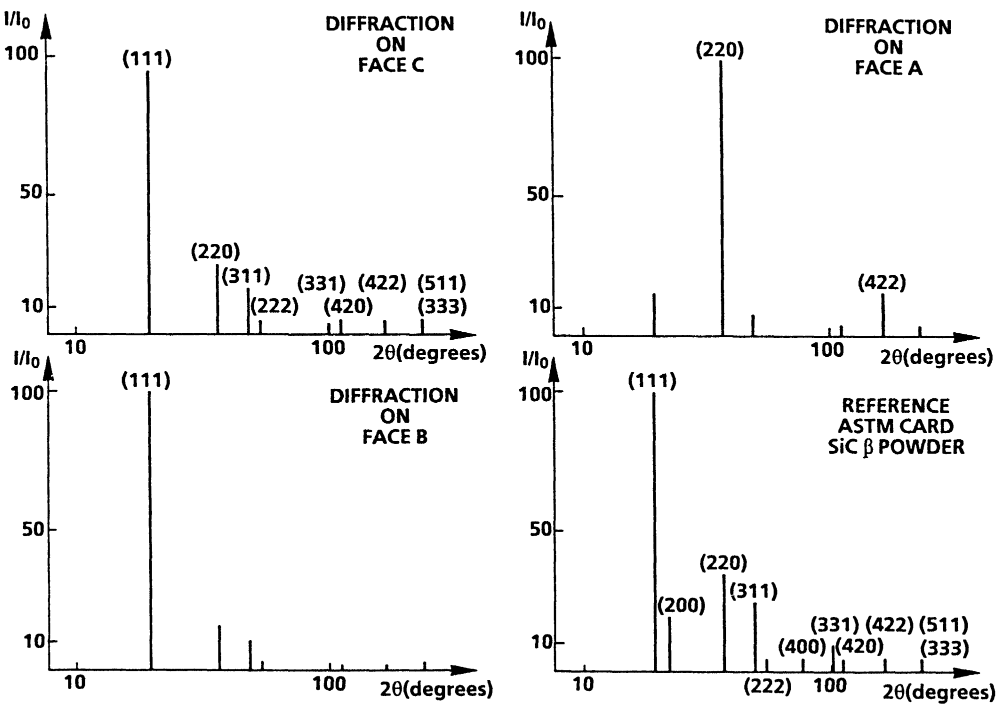

Figure 3 : Relative intensities vs. Bragg angle 2θ for ß-SiC (ASTM 29-1129) and for faces A, B and C of SiC/SiC composites.

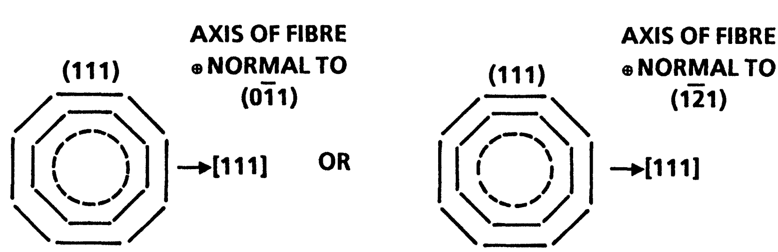

Figure 4 : Schematic representation of the SiC matrix texture. According to the stereographic projection of a cubic system, it is to be noted that the fibre axis can also be parallel to the [-1-2 3] direction of the matrix.

αβ goniometry

Several incomplete pole figures have been recorded with Schulz method[4] (in reflection) on face C of small unidirectional composite specimens (10 mm diameter and 2 mm thickness cylinders). No intensity correction have been performed. These pole figures, in particular the (111) and (220) pole figures (resp. fig. 5 and 6), confirm the results previously obtained with θ-2θ goniometry.

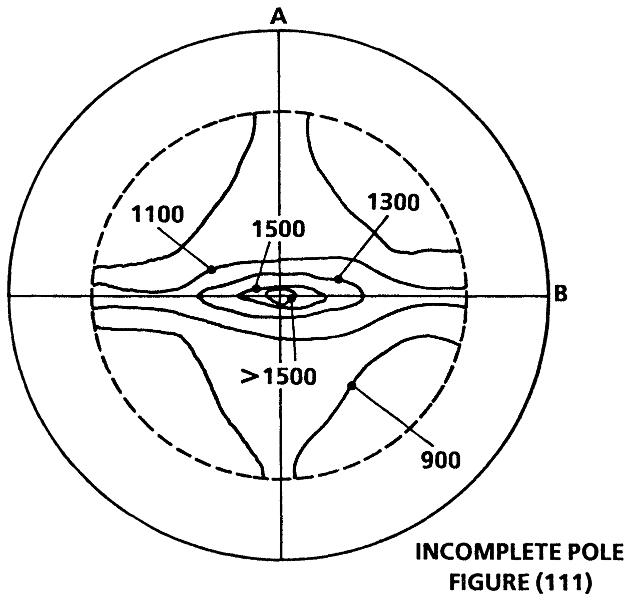

Figure 5 : Incomplete experimental (111) pole figure.

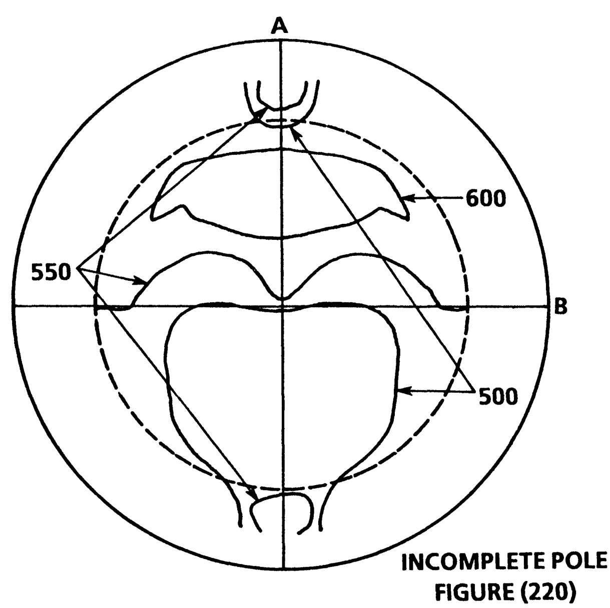

Figure 6 : Incomplete experimental (220) pole figure.

In figure 5, the equatorial line extending from B' to B is of the (111) type . One should expect for A, poles such as {110}, {112} or {123}. In figure 6, the richest region has a majority of poles making between 30 and 35° with BCB'; this confirms that the equatorial line corresponds to the {111} planes and A to a {112}-type plane. The other region which is rich in (220) poles is a narrow one located near A. A corresponds also to {110}-type planes.

3. DISCUSSION

The results presented clearly show that the SiC matrix exhibits a cylindrical texture with :

- {111} planes parallel to the fibre axis of the composite,
- {110} and {112} planes perpendicular to the fibre axis of the composite.

It has been noted by several authors (see for example ref. 5) that deposits formed by PVD or by CVD processes with low impinging atom fluxes tend to develop textures which place the lowest surface energy crystal facets parallel to the substrate; it is not therefore too surprising that under chemical vapour infiltration conditions adopted by SEP for face centered cubic ß-SiC, the deposit develops (111) planes parallel to the fibre surface.

The occurrence of the {110} and {112} components could simply arise from the preferred orientation of {111} planes : if (111) planes are parallel to the fiber axis, the planes containing [111] direction are perpendicular to this axis, this is the case for instance of (-1 1 0) and (-1-1 2) planes. There is no a priori physical reasons why the fibres should induce the deposit to grow with these planes perpendicular to their axis.

This texture may have significant consequences regarding the elastic constants of the SiC matrix of these materials. From the experimental values of compliances as determined by Li and Bradt[6], we have calculated Young's modulus for the main directions involved in the cylindrical texture. As no values seem to have been published for Young's modulus of ideally isotropic SiC material, the values determined according to Reuss and Voigt models are also given for comparison. Generally the values adopted in calculations for thermomechanical modelling range between 350 and 400 GPa[1,2]. The results, which are listed in table 1, show that Young's modulus value for [111] direction (511 GPa at RT) is significantly higher than the average values for isotropic materials.

Table 1 Young's modulus values calculated from experimental compliances published by Li and Bradt[6] (GPa)

T	$E_{<111>}$	$E_{<110>}$	$E_{<112>}$	$E_{<100>}$	E_{Reuss}	E_{Voigt}
RT	511	419	419	272	378	424
1000°C	489	397	397	254	357	405

It is not clear however what consequences these differences can have as far as thermal residual stresses or mechanical models are concerned, as no anisotropic calculations seem to have been carried for these composite materials; work is definitely needed in this area to assess this effect.
As an order of magnitude, it can be noticed that changing the Young's modulus value from 350 to 400 GPa increases the residual stresses in the matrix by 10%[2].

CONCLUSION

In the SiC/SiC ceramic unidirectional composites studied the silicon carbide matrix exhibits a cylindrical texture with {111} planes parallel to the fibre axis and with {110} and {112} planes perpendicular to the fibre axis. This texture arises from the chemical vapour infiltration process by which the matrix is formed.

Work is now under way to assess the effect of this texture on the thermomechanical behaviour of these composites.

References

1 - P. PERES : Analyse théorique et expérimentale du rôle des paramètres de microstructure sur le comportement des composites à matrice fragile. Thèse (INSA Lyon, 1988)

2 - V. ARNAULT : Relation entre microstructure et comportement mécanique des composites SiC/SiC : analyse du rôle de l'interface dans le processus de fissuration matricielle dans des matériaux multifilamentaires. Thèse (INSA Lyon, 1989).

3 - C. LAFFON et al. : Study of Nicalon-based ceramic fibres and powders by EXAFS spectrometry, X-ray diffractometry and some additionnal methods. *J. Mat. Sci.* **24** (1989) 1503-1512.

4 - H. BUNGE, C. ESLING : Quantitative Texture Analysis (H.J. Bunge and C. Esling edit.). DGM Informationsgesellschaft Verlag (1986).

5 - D.N. LEE : A model for development of orientation of vapour deposits. *J. Mat. Sci.* **24**(1989) 4375-4378.

6 - Z. LI, R.C. BRADT : The single-crystal elastic constants of cubic (3C) SiC to 1000°C. *J. Mat. Sci.*, **22** (1987), 2257.