INFLUENCE OF FIBER SURFACE PROPERTIES ON THE MECHANICAL BEHAVIOUR OF SIC/SIC MINICOMPOSITES

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Abstract (Times New Roman 12 pt, bold, single-line spacing, left-aligned text)
SiC/SiC composites reinforced with near stoichiometric SiC ceramic fibers (Hi-Nicalon S and SA3 Tyranno fibers) are attractive materials to be used as structural materials in fourth generation fission and fusion reactors.
The mechanical behaviour of CVI SiC/SiC composites with Hi-Nicalon S (HNS) or SA3 Tyranno (TSA3) fibers was investigated on minicomposites test specimen. Tensile tests have shown that the TSA3 fiber reinforced composites exhibited a brittle behaviour whereas composites reinforced by HNS showed non brittle tensile curve. These differences may be attributed to the fiber surface properties, which influence the fiber-matrix interface. Fiber surface morphology and chemical extreme surface have been compared for both fibers. Results show that fiber surface roughness of TSA3 is the main characteristic which could explain the brittle behaviour of materials.

1 Introduction

SiC/SiC composites are commonly used for high-temperature engineering applications due to their excellent specific mechanical properties at elevated temperatures. Their good microstructural stability under high-energy neutron irradiation makes them attractive to be used as blanket components in fusion or structural material in fission reactors [1-5]. In the frame of nuclear applications, their mechanical properties remain however a drawback and must be adjusted to resist to neutronic irradiation. This mechanical behaviour of the composite depends on the fiber/matrix (F/M) bonding. Among the parameters influencing the mechanical properties of the composite, the nature of the C/SiC interface and/or the surface characteristics of the SiC fibers appear to be predominant. The SiC surface characteristics depend on the fibers type [13-16]. Two fibers types appear interesting to be used in SiC/SiC composites devoted to neutron irradiation: the fibers named Hi-Nicalon S and SA3 Tyranno [6, 7]. The mechanical behaviour of CVI SiC/SiC composites reinforced with Hi-Nicalon S (noted HNS) or SA3 fibers (noted TSA3) were investigated on minicomposite (unidirectional composite reinforced with single tows) test specimen. Tensile tests and push-out tests were done in order to determine the interfacial shear strength. The surface properties of SiC fibers were investigated in relation with the mechanical behaviour of the corresponding composite in order to identify the predominant surface characteristics responsible for the mechanical

behaviour. The surface morphology and the chemical characteristics were determined using several characterization techniques. The results are discussed in order to highlight the importance of the SiC surface properties on the F/M coupling.

2 Materials and testing methods

2.1 Materials

SiC/SiC composites were prepared from unidirectional materials (minicomposites) constituted of fibers, pyrocarbone interphase (e ~ 150nm) and SiC matrix. The interphase and the matrix were elaborated by Chemical Vapor Infiltration (CVI).

Two SiC fibers were characterized in terms of morphology, surface roughness and surface chemistry. Prior to the fibers analysis, the sizing was removed by heat treatment at 900°C under vacuum (pressure ~ 10^{-4} Pa) for one hour. These parameters are close to the one used during the fabrication of the macroscopic CVI SiC/SiC composite.

2.1 Testing methods

2.2.1 Characterization of SiC fibers

The surface roughness of the fibers was determined from Atomic Force Microscopy images in tapping mode. Images of 500nm x 500nm were taken for all samples.

X-ray Photoelectron Spectroscopy (XPS) was used to study the surface chemistry of the SiC fibers. The analysed surface area was 0.5mm x 6mm. A survey spectrum and high-resolution spectra of carbon (C1s peak), oxygen (O1s peak) and silicon (Si2p peak) were recorded.

2.2.1 Mechanical tests

Tensile tests were performed on SA3 Tyranno and Hi-Nicalon S reinforced composites at room temperature at a constant rate (50µm/min). Uniaxial tension tests used a 500 N load cell.

Single-fiber push-out tests were performed on the SiC/SiC minicomposite specimen test using a nanoindenter to measure the intensity of the fiber/matrix bond and the interfacial shear stress. The samples were observed using SEM to check if the fibers are correctly loaded and the absence of damage on the fibers and matrix during the tests.

3 Results and discussion

The stress-strain curves of the minicomposites reinforced with HNS and TSA3 fibers at room temperature are reported in Figure 1.

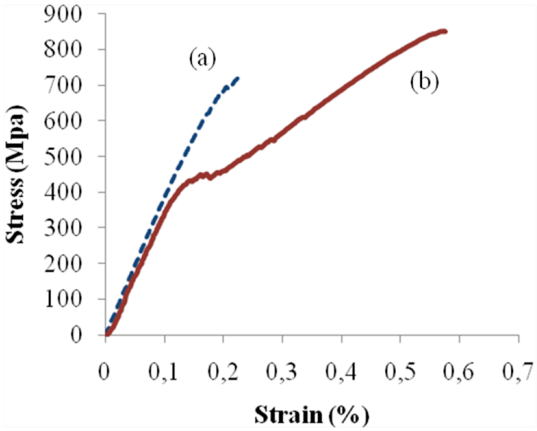

Figure 1. Typical behaviour obtained with (a) the SA3 Tyranno and (b) Hi-Nicalon S reinforced SiC/SiC minicomposites.

Both composites show a typical behaviour [8-10]. A difference in the tensile strain is however observed since the failure strain of the composites reinforced by TSA3 fibers is smaller (ε_r ~ 0.3%) than the one of the HNS reinforced composites (ε_r ~ 0.7%). The different mechanical behaviour observed can be attributed to the F/M interfacial coupling. The mechanical

properties of the F/M interface were measured thanks to the push-out test which emerged as the standard test [11-13]. Single-fiber push-out tests were carried out on the SiC/SiC minicomposites. The analysis of the load - displacement curves gives access to the strength (F_g) that must be applied to the fiber to extract itself in totality and hence to disrupt the interfacial bonding. The measurement of F_g allows to calculate the interfacial shear strength (τ_s) using the following equation (Eq. (1)):

$$\tau_s = \frac{F_g}{2.\pi.r_f.l_f} \quad (1)$$

Where r_f is the fiber radius and l_f the fiber length.

Material	HNS/SiC	TSA3/SiC
τ_s (MPa)	15	109

Table 1. Interfacial shrear strength obtained by push-out tests on SiC/SiC composites using HNS and TSA3 fibers.

As seen in Table 1, the interfacial shear strength is much higher for the TSA3-minicomposites. It indicates that the F/M bonding is stronger when the composite is reinforced with TSA3 fiber compared to HNS fiber as already suggested by the strain failure value. Since both composites only differ by the nature of their reinforced fibers, the difference in the strength of the F/M interfacial bonding can be related to the fiber/interphase bonding linked to the fiber surface properties.
To better understand the influence of surface properties, both types of fibers will be analyzed in terms of surface roughness and surface chemistry as described in the following section.

3.1 Surface roughness of SiC fibers
Fibers surface roughnesses were quantified by AFM in tapping mode. The AFM images of the fiber surface are reported in Fig. 2. The values of R_{max} and R_{RMS} summarized in table 2 correspond to the maximum amplitude determinated in the analyzed surface (500nm x 500nm) and to the quadratic mean amplitude, respectively.

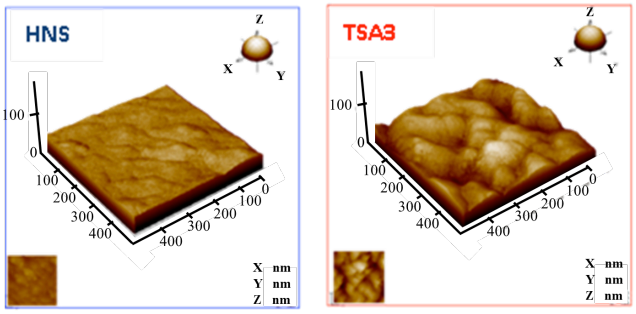

Figure 2. AFM images of HNS and TSA3 fibers.

Fiber	HNS	TSA3
R_{RMS} (nm)	3.0	9.0
R_{max} (nm)	25.0	56.1

Table 2. Results of fibers surface roughness

It can be noted that the R_{RMS} value is three times higher for TSA3. The surface roughness discrepancy between the two types of fibers is significant enough to modify the interfacial

shear strength (τ_s). Shear stress, determinated by the push-out test (Eq. (1)), is also linked to the surface roughness as explained below. In fact τ_s can be calculated as follows:

$$\tau_s = -\mu\sigma_T \tag{2}$$

where μ is the friction coefficient and σ_T the residual stresses.

Furthermore, the radial residual stresses (σ_T) can be determined as follows (Eq. (3)).

$$\sigma_T = \sigma_{th} + \sigma_R \tag{3}$$

Where σ_R corresponds to the radial residual stress induced by fiber surface roughness and σ_{th} is the thermally radial residual stress which results from the mismatch in the thermal expansion coefficient between the fiber and the matrix. The values of σ_{th} in radial direction for both types of fibers are reported in Table 3.

Fiber	HNS	TSA3
σ_{th} (MPa)	-200	-130
σ_R (MPa)	184	1020
σ_T (MPa)	-16	890

-= tensile and += compressive

Table 3. Radial residual stresses induced by: thermally radial residual stresses (σ_{th}) [10], fibers surface roughness (σ_R) described by Eq. (5) and computation of radial residual stresses ($\sigma_T = \sigma_{th} + \sigma_R$).

They were determined as explained in our previous paper by considering a single fiber and concentrically cylinders of fiber and matrix [10].
It can be noted that the values of σ_{th} are close for both fibers. As a consequence, σ_T in radial direction will mainly depend on σ_R which is related to the surface roughness according to the following equation:

$$\sigma_R = (\frac{A}{r_f})\gamma \tag{4}$$

with $A = 2 * R_{RMS}$

where A is the amplitude of lateral displacements induced by the surface roughness quantified through the RMS parameter and γ, mechanical characteristics of SiC fibers.
As indicated in Table 3, the values of σ_{th} and σ_R compensate themselves (compressive and tensile radial stresses are close) for the HNS-based minicomposite leading to a low value of the radial residual stresses (σ_T). On the contrary, the TSA3 minicomposite exhibits a much higher radial residual stresses due to the high value of the compressive radial stresses induced by the important surface roughness of the TSA3 fiber. As a consequence the τ_s discrepancy observed between the two types of fibers can be attributed to the surface roughness difference. These results confirm that the surface roughness is one of the main parameter which controls the interfacial shear stress and consequently the F/M coupling [14].

3.1 Surface chemistry of SiC fibers
The atomic concentration of C, O and Si deduced from the survey XPS spectra analysis are summarized in Table 4.

Fiber type	C1s (at.%)	O1s (at.%)	Si2p (at.%)
HNS	83.9	5.6	10.6
TSA3	84.3	4.1	11.6

Table 4. XPS qualitative analysis of Hi-Nicalon S (1), Hi-Nicalon S (2) and SA3 Tyranno fiber

The survey analysis confirms that the surface fiber is mainly composed of carbon (~ 84%) whatever the fibers type. To identify the nature of chemical bonds, high resolution spectra of C1s, O1s and Si2p were recorded and fitted. Resulting data for HNS and TSA3 fibers are summarized in Table 5.

Fiber		HNS	TSA3
Components			
C1s (%)	C-Si	7.3	8.2
	Csp_2	71.5	68.7
	Csp_3	12.6	16.0
	C-CO	0.8	0.6
	C-O	0.8	0.2
	SiO_xC_y	2.5	1.7
	C=O	0.7	1.7
	COOR	0.8	0.6
Si2p (%)	Si-C 3/2	31.7	45.2
	Si-C 1/2	31.7	45.2
	$SiOC_3$ 3/2	0	0.1
	$SiOC_3$ ½	0	0.1
	SiO_2C_2 3/2	4.5	3.5
	SiO_2C_2 ½	4.5	3.5
	SiO_3C 3/2	3.2	0.8
	SiO_3C 1/2	3.2	0.8
	SiO_2 3/2	10.5	0.5
	SiO_2 1/2	10.5	0.5

Table 5. Results of high resolution spectra fitted for HNS and TSA3 fibers

Sp^2 bonded carbon is mainly present on the surface of HNS and TSA3 fibers. A small proportion of aliphatic carbon is observed for two SiC fibers. This carbon layer on SiC fibers is important to promoted C-C bonds with pyrocarbone interphase. Moreover, the decomposition of the Si2p spectra reveals that SiC, SiO_xC_y and SiO_2 are also present on the fiber surface (Table 5). Their atomic concentration remains however low compared to the carbon fraction. The TSA3 surface is mainly composed of SiC; the fraction of SiO_2 being negligible. On the contrary, the SiO_2 fraction is much higher in the case of the HNS fiber. The presence of SiC and SiO_2 is not significant enough to modify the nature of the fiber/matrix interphase bond and consequently the mechanical properties.
The experimental results point out that the quality of the F/M interface responsible of the mechanical properties will depend on the fiber surface roughness and fiber surface chemistry.

4 Conclusion

Two types of SiC fibers were analyzed (TSA3 and HNS). As highlighted by the experimental results, the mechanical behaviour of the minicomposite is correlated to the surface roughness and surface chemistry of the SiC fibers. Both parameters were quantitatively determined by AFM and XPS. The analysis point out that the TSA3 and HNS fibers display different surface characteristics. Contrary to the HNS fibers, TSA3 has a granular and rough surface which leads to an increase of the residual stress and consequently the interfacial shear strength. The surface of both fibers is mainly composed of sp^2 bonded carbon. As a result, the F/M bonding

will be stronger in the case of the TSA3-based minicomposite explaining the more brittle behaviour. Therefore, this work clearly points out that the strength of the F/M interfacial bonding is controlled by the surface roughness and surface chemistry of SiC fibers.

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