THERMOMECHANICAL BEHAVIOUR OF 2D-SiC/SiC COMPOSITES WITH NANOSCALE-MULTILAYERED $(PyC/SiC)_n$ INTERPHASES

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SUMMARY: 2D woven SiC/SiC composites, with nanoscale-multilayered $(PyC/SiC)_n$ interphases, have been made via chemical vapour infiltration of a preform of either Nicalon or Hi-Nicalon fibres. The nanoscale-multilayered $(PyC/SiC)_n$ interphases consist of n successive PyC-SiC sequences. In the present paper n = 10 and the thickness of the PyC and SiC sublayers was about 20 nm and 50 nm respectively. The influence of Hi-Nicalon fibres and multilayered interphases on the mechanical behaviour at room temperature and the lifetime in cyclic fatigue at 600°C and 1200°C was investigated. The Hi-Nicalon fibre and the multilayered interphases are shown to lead to improved 2D-SiC/SiC composites.

KEYWORDS: interphase, mechanical properties, lifetime, cyclic fatigue, oxidation, ceramic matrix composite, multilayer.

INTRODUCTION

Ceramic Matrix Composites (CMC) are potential candidates for high temperature structural applications. The pyrocarbon (PyC) interphase, which is commonly used to monitor the fibre/matrix interactions in SiC/SiC composites, oxidizes at high temperature. Recently, the concept of $(PyC/SiC)_n$ multilayered interphase has been developed [1]. First, microscale-multilayered $(PyC/SiC)_n$ interphases in 2D woven Nicalon/SiC composites were prepared according to the I-CVI route [2]. Then, nanoscale-multilayered $(PyC/SiC)_n$ interphases, deposited by pulsed-CVD, were introduced into Nicalon/SiC microcomposites [3]. More recently, $(PyC/SiC)_n$ and $(BN/SiC)_n$ nanoscale-multilayered interphases, deposited by pulsed-CVI, were introduced into SiC/SiC minicomposites, each reinforced with a single Hi-Nicalon fibre tow [4].

The present paper investigates the thermomechanical behaviour of 2D woven SiC/SiC composites with nanoscale-multilayered $(PyC/SiC)_n$ interphases.

EXPERIMENTAL

Materials

The SiC/SiC composites were reinforced by treated fabrics of either NLM 202 Nicalon fibres (from Nippon Carbon, Japan) and Hi-Nicalon fibres (from Nippon Carbon, Japan).

The interphases were deposited via the pressure-pulsed CVI process [5], whereas the SiC-matrix was made by the classical I-CVI route (by S.E.P., Le Haillan, France). Composites with the following interphases were investigated : either a single pyrocarbon interphase (100 nm thick, referred to as $(PyC_{100}/0)_1$) or a nanoscale-multilayered interphase $(PyC_{20}/SiC_{50})_{10}$, consisting of 10 PyC-SiC sequences (the thickness of each sublayer was respectively $e_{(PyC)}$ = 20 nm and $e_{(SiC)}$ = 50 nm) (Table 1). The fibre volume fraction was about 40% and the residual porosity was about 10-15%.

Table 1: Batches of 2D-SiC/SiC composites which have been manufactured.

Batches	Fibres	Interphase	Interphase parameters		
			$e_{(PyC)}$ (nm)	$e_{(SiC)}$ (nm)	n
A	Nicalon NLM 202	$(PyC_{20}/SiC_{50})_{10}$	20	50	10
D	Nicalon NLM 202	$(PyC_{100}/0)_1$	100	0	1
F	Hi-Nicalon	$(PyC_{100}/0)_1$	100	0	1
G	Hi-Nicalon	$(PyC_{20}/SiC_{50})_{10}$	20	50	10

Microstructural and mechanical characterization

The $(PyC/SiC)_n$ interphases were examined by scanning electron microscopy (SEM).

Tensile tests were performed at room temperature, at a constant strain rate of $0.1\%.min^{-1}$. The deformations were measured using an extensometer (gauge length = 25 mm). The tests specimens (dimension : 200 mm *10 mm * 3 mm) have been prepared from rectangular plates. The interfacial shear stress (τ) was estimated from the area of the hysteresis loops observed during unloading-reloading cycles [6].

Tensile cyclic fatigue tests were carried out at high temperature, in air, under load-control using a sinusoidal wave-form with a frequency of 0.25 Hz and a stress ratio $R = \sigma_{min} / \sigma_{max} = 0.1$. The maximum stress ($\sigma_{max}$), that was applied, was 120 MPa. Two temperatures were examined : 600°C and 1200°C.

RESULTS

Interphase characterization

Fig. 1 shows a SEM image of the nanoscale-multilayered $(PyC_{20}/SiC_{50})_{10}$ interphase. The PyC and SiC sublayers form concentric cylinders around the fibres, as already observed for the nanoscale-multilayered $(PyC/SiC)_n$ interphases in SiC/SiC minicomposites [4] and microcomposites [3]. The SiC sublayers exhibit a rough aspect at the nm-scale. Nanocristallization of the SiC grains was achieved through appropriate processing conditions [3-5].

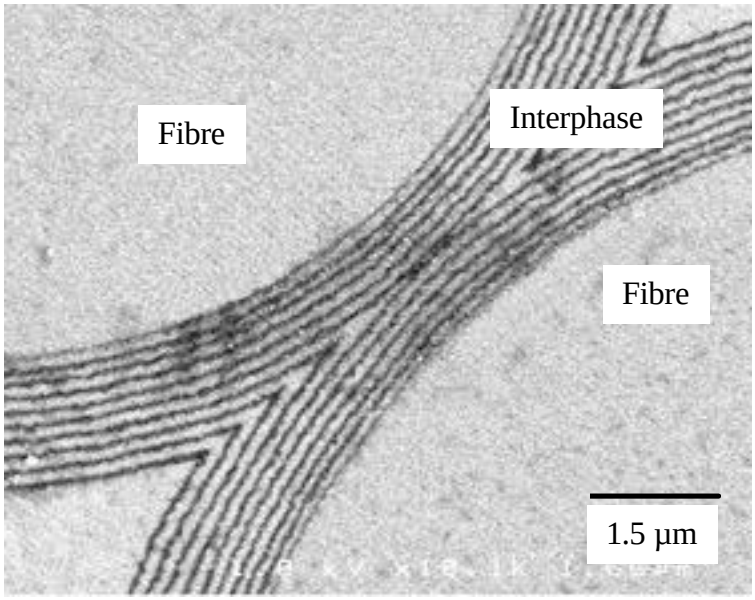

Fig. 1: SEM micrograph of the nanoscale-multilayered $(PyC_{20}/SiC_{50})_{10}$ interphase in a 2D-SiC/SiC composite (dark : PyC sublayer ; white : SiC sublayers).

Mechanical behaviour at room temperature

The tensile stress-strain curves (σ-ε) shown in Fig. 2, exhibit a significant non-linear domain, indicative of a non brittle response. The features of the stress-strain behaviour including Young's modulus, proportional limit and resistance-to-failure are given in Table 2. The influence of reinforcing fibres and multilayered interphases can be noticed from Table 2 and Fig. 2. Thus the composites reinforced with Hi-Nicalon fibres display the larger characteristics, except the resistance-to-failure for batch F. Furthermore, those composites with a multilayered interphase exhibit larger features than their counterpart with a single layer interphase.

These trends are in agreement with the estimates of the interfacial shear stress τ, which reflect stronger fibre/matrix interactions in those composites reinforced with Hi-Nicalon fibres and with multilayered interphases (except for specimens of batch F which probably experienced a premature failure). However, the interfacial shear stresses reflect rather strong fibre/matrix interactions through the debonded interphases [2] for all the batches, except for batch F. For batch F, it may be excepted that the appropriate interfacial shear stress could not be determined because of the premature failure of specimens.

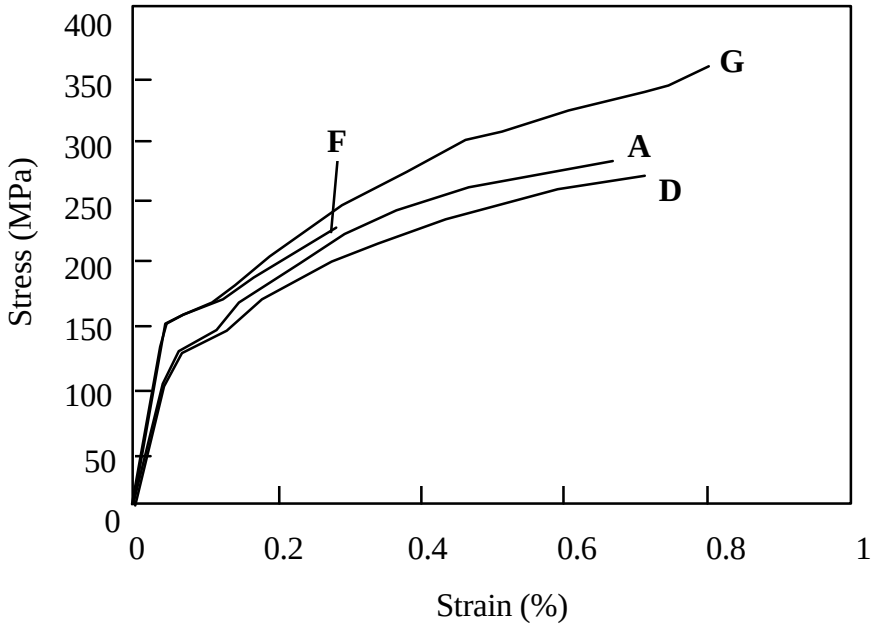

Fig. 2: Tensile stress-strain curves for the 2D-SiC/SiC composites with nanoscale-multilayered $(PyC/SiC)_n$ interphases (A and D : composites reinforced with treated Nicalon fibres ; F and G : composites reinforced with treated Hi-Nicalon fibres).

Table 2: Mechanical characteristics of the 2D-SiC/SiC composites.

Batches	Young's modulus	Proportional limit		Failure		τ
	E (GPa)	σ (MPa)	ε (%)	σ (MPa)	ε (%)	(MPa)
A	249	90	0.036	273	0.68	190
D	274	100	0.037	258	0.71	167
F	322	128	0.040	222	0.29	66
G	316	133	0.042	346	0.81	232

Lifetime

The average values of lifetime which were obtained for each batch at 600°C and 1200°C are plotted in Fig. 3. An improvement of the lifetime is provided by the nanoscale-multilayered interphase, as shown by the comparison of either batches A and D, or batches G and F. A more significant improvement is obtained with the Hi-Nicalon fibres, as shown by the comparison of either batches G and A or batches F and D. The influence of multilayered interphases and Hi-Nicalon fibres seems to be less significant at 1200°C, except for batch G. Specimens of batch F did not display the shortest lifetime, which suggests that the ultimate failure was premature at room temperature.

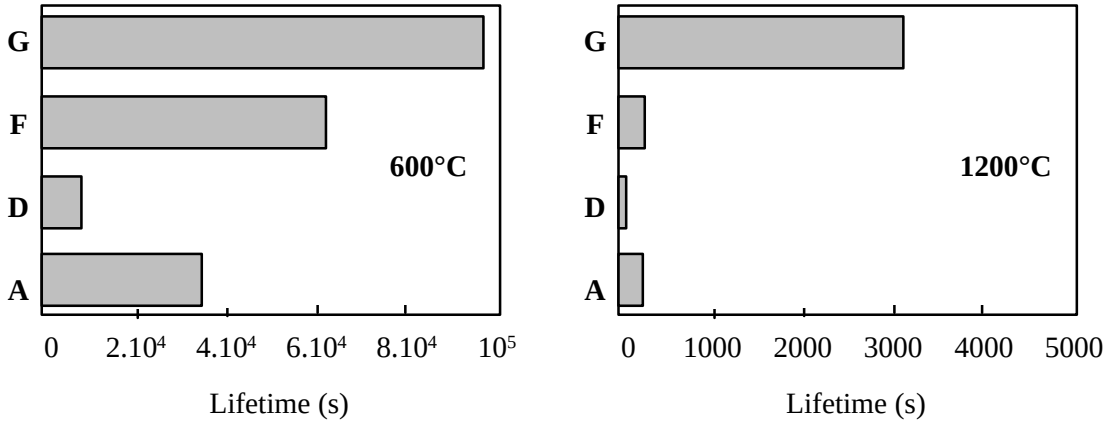

Fig. 3: Lifetime of the 2D-SiC/(PyC/SiC)$_n$/SiC composites at 600°C and 1200°C, in air, under cyclic fatigue conditions.

DISCUSSION

Influence of fibre properties

A significant difference between the Nicalon and Hi-Nicalon fibres lies in the Young's modulus : 200 GPa for the Nicalon fibres against 305 GPa for the Hi-Nicalon fibres [7]. This discrepancy influences the Young's modulus of the composite as well as the proportional limit. The higher proportional limit exhibited by the Hi-Nicalon/SiC composites must be attributed to the presence of lower stresses in the matrix, as a result of the fibre Young's modulus which improves the load sharing.

The difference in coefficients of thermal expansion (α_L) must also be considered : α_L = 3.910^{-6} °C^{-1} for the Nicalon fibres and α_L = 4.610^{-6} °C^{-1} for the Hi-Nicalon fibres [7]. The coefficient of thermal expansion of the matrix α_m = 4.610^{-6} °C^{-1} [7], is similar to that of Hi-Nicalon fibres. Therefore, negligible residual stresses may be expected in the matrix of the Hi-Nicalon/SiC composites after cooling from the processing temperature. On the contrary, in the Nicalon/SiC composites, tensile residual stresses are induced in the matrix as a result of the thermal expansion mismatch. These residual stresses add to the magnitude of the stresses operating on the matrix, which favours the onset of matrix cracks, and, consequently, affects the proportional limit.

It is known that at high temperature, the oxidation resistance of the Hi-Nicalon fibres is improved over the Nicalon fibres [8]. Therefore, it seems logical to relate the effect of Hi-Nicalon fibres on the lifetime, to their oxidation resistance. However, further investigation of the oxidation of Hi-Nicalon fibres is necessary to refine the analysis of the lifetime of the Hi-Nicalon/SiC composites.

Influence of interphases

The tensile tests at room temperature indicate that multilayered interphases tend to increase the interfacial shear stress τ. This effect may be attributed to the deviation of matrix cracks into multiple cracks in the PyC sublayers [2-5]. This phenomenon tends to decrease the

debond length and also to increase the fibre/matrix interactions through the debond [2]. A comparable influence of multilayered interphases on the interfacial shear stress τ has been noted in [9].

At high temperature, the presence of multiple cracks in the PyC sublayers [5] is expected to increase the oxygen diffusion paths and therefore to slow down the oxidation of the PyC interphases [4,10] thus leading to longer lifetimes.

CONCLUSIONS

2D-SiC/SiC composites with nanoscale-multilayered $(PyC/SiC)_n$ interphases reinforced with treated Nicalon or Hi-Nicalon fibres have been made via the chemical vapour infiltration process. The investigation of these composites has demonstrated that the nanoscale-multilayered $(PyC/SiC)_n$ interphases deposited by P-CVI can replace the single PyC interphase. A similar conclusion had been reached with *(i)* microscale-multilayered $(PyC/SiC)_n$ interphases deposited by I-CVI, in 2D Nicalon/SiC composites [2] and *(ii)* nanoscale-multilayered $(PyC/SiC)_n$ interphases deposited by P-CVI, in Hi-Nicalon/SiC minicomposites [4] or Nicalon/SiC microcomposites [3].

The stress-strain behaviour, in tension at ambient temperature, was improved by the nanoscale-multilayered interphases. Furthermore the interfacial shear stress was increased. It characterizes rather strong fibre/matrix interactions through the debond. The Hi-Nicalon fibres increase the mechanical properties of the composites as a result of their high Young's modulus and the absence of thermal expansion mismatch between fibre and matrix.

The oxidation resistance of the composites is controlled by two factors : the oxidation resistance of the fibres and that of the interphase. Hi-Nicalon fibres and multilayered $(PyC/SiC)_n$ interphases increase the lifetime of composites.

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REFERENCES

1. Naslain, R., "The concept of layered interphases in SiC/SiC", *Proceedings of the Second International Conference in High Temperature Ceramic Matrix Composites (HT-CMC II)* : Manufacturing and Materials Development, *Ceram. Trans.*, Santa Barbara, USA, August 21-24, 1995, Vol. 58, Evans, A.G. and Naslain, R., Eds., pp. 23-26.

2. Droillard, C. and Lamon, J., "Fracture toughness of 2D vowen SiC/SiC CVI-composites with multilayered interphases", *J. Amer. Ceram. Soc.*, Vol. 79, 1996, p. 849.

3. Heurtevent, F., "Nanoscale multilayered $(PyC/SiC)_n$ materials - Application as interphases in CMCs", Ph. D. Thesis, No. 1476, March 29, 1996, University of Bordeaux, France (in French).

4. Bertrand, S., Boisron, O., Pailler, R., Lamon J. and Naslain, R., "$(PyC/SiC)_n$ and $(BN/SiC)_n$ nanoscale-multilayered interphases by pressure pulsed-CVI", *Proceedings of the Third International Conference on High Temperature Ceramic Matrix Composites (HT-CMC III), The Ceramic Society of Japan. (CSJ series, Vol. 3), Key Engineering Materials*, September 6-9, 1998, Osaka, Japan, Niihara, K. Nakano, K., Sekino, T. and Yasuda, E., Eds., 1999, Vols. 164-165, pp. 357-360.

5. Bertrand, S., Germain, F., Pailler, R., Lamon, J. and Goujard, S., "Study of 2D-SiC/SiC composites with nanoscale-multilayered $(PyC\text{-}SiC)_n$ interphases", submitted to *J. Amer. Ceram. Soc.*.

6. Lamon, J., Rebillat, F. and Evans, A.G., "Microcomposite test procedure for evaluating the interface properties of ceramic matrix composites", *J. Amer. Ceram. Soc.*, Vol. 78[2], 1995, pp. 401-405.

7. Bertrand, S., Forio, P., Pailler, R. and Lamon, J., "Hi-Nicalon/SiC minicomposites with $(PyC\text{-}SiC)_n$ nanoscale-multilayered interphases", *J. Amer. Ceram. Soc.*, in press.

8. Chollon, G., Pailler, R., Naslain, R., Laanani, F., Monthioux, M. and Olry, P., "Thermal stability of a PCS-derived SiC fiber with a low oxygen content (Hi-Nicalon)", *J. Mater. Sci.*, 1997, Vol. 32, p. 327.

9. Rebillat, F., Lamon, J., Naslain, R., Lara-Curzio, E., Ferber, M.K., and Besmann, Th.M., "Properties of multilayered interphases in SiC/SiC chemical vapor infiltrated composites with ‘weak’ and ‘strong’ interfaces", *J. Amer. Ceram. Soc.*, Vol. 81[9], 1998, pp. 2315-26.

10. Pasquier, S., Lamon, J. and Naslain, R., "Tensile static fatigue of 2D-SiC/SiC composites with multilayered $(PyC/SiC)_n$ interphases at high temperature in oxidizing atmosphere", *Composites-Part A*, Vol. 29A, 1998, pp. 1157-1164.