Journal of the European Ceramic Society **18** (1998) 1827–1833

0955-2219/98/$—see front matter
PII: S0955-2219(98)00122-8

Apparent Stiffening of Ceramic-matrix Composites Induced by Cyclic Fatigue

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Abstract

This work carries on four different long-fibre-reinforced ceramic-matrix composites: a cross-weave SiC/SiC, a cross-ply SiC/MAS-L, a cross-weave C/SiC and a $[0,+60,-60]_n$ *C/C laminate. Experimentally, cyclic fatigue effect has been observed at room temperature, at high temperature under inert atmosphere, and at room temperature after a previous ageing at high temperature under vacuum. For these four materials, the evolutions of the macroscopic mechanical behaviour with the number of cycles applied can be explained by an evolution of interfaces as well, fibre/matrix interfaces as neighbouring ply interfaces, according to the following mechanisms: (i) interfacial wear of interfaces due to to-and-fro sliding of fibres or of plies under cyclic loading, and (ii) dependence of the residual thermal stresses with the temperature of the test. Previous ageing at high temperature under vacuum can also enable in CMC some physical and chemical changes in the constituents leading for example to a slight removing of fibre/matrix interphases by oxidation. Usually, damage induced by cyclic fatigue in long-fibre-reinforced ceramic-matrix composites leads to a reduction of the tensile apparent elastic modulus as cycling proceeds. But an original macroscopic stiffening has been experimentally observed during cyclic fatigue. This phenomenon has been observed on C/C composite at room temperature, on C/SiC and on SiC/MAS-L at high temperature, and on SiC/SiC at room temperature after previous ageing under vacuum at high temperature. This apparent stiffening is not well understood at present time, but appeared in materials with low interfacial shear strength and is seemingly due to incomplete closure during unloading of the cracks present in transverse yarns.*

1 Introduction

Under cyclic loading, the mechanical behaviour of long-fibre-reinforced ceramic-matrix composites changes progressively as a function of the number of cycles and can lead to fatigue failure.[1–7] The main mechanism involves matrix cracking in the longitudinal and transverse yarns on the first loading cycle, depending on the detail of the architecture of the reinforcement. This cracking is followed by a debonding and a cyclic sliding along fibre/matrix or yarn/yarn interfaces. Repeated sliding causes wear phenomena at the interfaces, leading to a reduction of the stress transfer capability and a corresponding increase of the fibre failure probability, i.e. a decrease of fibre-bundle strength and of fibre-bundle stiffness. The connection between wear and fatigue effects was first pointed out in Refs[2,3]. In addition, there is a close relationship between the level of interfacial shear stress (ISS) and the width of the stress-strain loops (or the relative energy dissipated per cycle called mechanical hysteresis). In the saturated matrix cracking situation, the loop becomes less and less open as the ISS decreases. Conversely, the non-saturated matrix cracking case leads to an increasing width of the loops as the ISS decreases.[2,7]

On the macroscopic mechanical behaviour, many parameters can be defined in order to describe quantitatively the shape of the hysteresis loops observed under cyclic fatigue, and its evolution during a cyclic fatigue test. We can, for example, follow the evolution of the mean elastic modulus (which is the mean slope of the stress/strain loops), the mechanical hysteresis (corresponding to the area of the stress/strain loop), and the residual strain (strain at zero load) (see Fig. 1).

Concerning the mean elastic modulus we defined in order to characterize the shape of the stress/strain loops observed during cyclic fatigue, care should be taken about its meaning. In long-fibre-reinforced ceramic-matrix composites, the true elastic modulus (Young's modulus) is measured with cycles of small amplitudes whatever level of stress. In that case, the modulus is only given by the elastic strains of the constituents of the composite, without any sliding between constituents. But under cycles of big amplitudes, slidings between constituents occur in the composite, and

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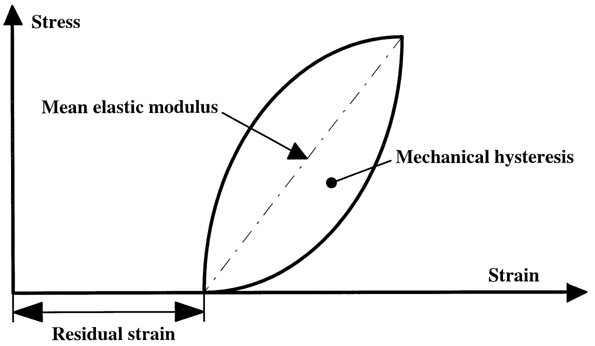

Fig. 1. Definition of the residual strain (strain at zero loading), of the mean elastic modulus (slope of the stress/strain loop) and of the mechanical hysteresis (area of the loop), to characterize the shape of stress/strain loops observed during cyclic fatigue tests.

the shape of the stress/strain loops include the elastic strains and the non-elastic strains. Hence, the mean elastic modulus which is the mean slope of the stress/strain loop is not representative of the elastic behaviour of the composite, but is representative of its macroscopic stiffness.

In materials, the mean elastic modulus usually decreases during cyclic fatigue as long as damage extends in the material. This parameter is commonly used to quantify the amount of damage created in a material under mechanical loading. This is the classical damage parameter. In addition the mechanical hysteresis increases as long as the material is damaged, and the residual strain increases too in correlation. But in long-fibre-reinforced ceramic-matrix composites these three parameters can evolve differently, because of specific micromechanisms of damage. For example, concerning the mechanical hysteresis, this parameter can increase then decrease after reaching a maximum value.[7] Concerning the mean elastic modulus we have pointed out an apparent stiffening observed in many long-fibre-reinforced ceramic-matrix composites, as well at high temperature under inert atmosphere as at room temperature with or without a previous heat-treatment under vacuum.

The aim of this paper is to present this nonclassical stiffening observed during cyclic fatigue in four composites presenting complex reinforcement architecture: C/C, SiC/MAS-L, SiC/SiC and C/SiC.

2 Materials studied

The experimental studies carried on four different long-fibre-reinforced ceramic-matrix composites: a C/C composite, a SiC/MAS-L composite, a SiC/SiC composite and a C/SiC composite.

In this study, the C/C composite is a $[0, +60, -60]_n$ laminate provided by Carbone Industrie and the Société Européenne de Propulsion (S.E.P.). Each ply is made from parallel carbon yarns embedded in carbon matrix. The SiC/MAS-L composite is a cross-ply symetrical composite with plies made from SiC Nicalon fibres embedded in glass matrix. This material is provided by Aérospatiale. The SiC/SiC composite is a cross-weave composite made from SiC Nicalon fibres embedded in SiC matrix deposited by chemical vapour infiltration. This so called 2D SiC/SiC GS4C is not protected against oxidation. This material is provided by S.E.P., and, with the SiC/MAS-L, was studied within the frame of the scientific association: 'Thermomechanical behaviour of fibrous ceramic–ceramic composites'. The C/SiC composite is also provided by S.E.P. It is a cross-weave composite made from carbon fibre embedded by C.V.I. SiC matrix. Like the SiC/SiC material, this material is not coated against oxidation.

3 Experimental procedure

The four materials presented in this paper have been tested under uniaxial tensile or compression loading with the axis of loading parallel to a direction of fibres reinforcement. The experiments were performed on two hydraulic servo-controlled machines: a MTS 810 device, and a Instron 8502 device fitted out with an inductive furnace. The MTS system was used for all the tests at room temperature. The Instron system is able to perform tests from room temperature to 1600°C under air, and to 2000°C under vacuum (10^{-2} mbar), or under inert atmosphere (argon) with a servo-controlled pressure from 1 mbar to atmospheric pressure. This device was used in this study to perform the tests at high temperature up to 1500°C under 30 mbar of argon.

The specimens were classical tensile specimens with a uniform gauge length of 45 mm. Some specimens have been previously aged at high temperature under vacuum with the Instron system. For these particular specimens, the cyclic fatigue tests were then performed at room temperature on the MTS system.

All the specimens have been subjected to a sinusoïdal servo-controlled variation of the load at a frequency of 1 Hz as well for tensile/tensile tests as for tensile/compression tests.

4 Results and discussion

As described above, the specimens were tested at constant amplitude and at constant frequency.

When the materials are tested under those conditions they describe a loop in the stress/strain diagram. The evolution of this loop is experimentally followed during the test, and the changes in shape with the number of applied cycles are observed on the stress/strain diagram or on other diagrams like mean tension elastic modulus/number of cycles for example. In order to characterize the shape of the stress/strain loops, three parameters can be used: the mean tension elastic modulus, the mechanical hysteresis (area of the stress/strain loop) and the residual strain.

Figure 2 shows typical loops observed during cyclic fatigue at room temperature of the SiC/SiC composite. It is clearly shown, in the tensile part, that the loops become more open and less stiff as fatigue proceeds. On the other hand, the material remains elastic in the compressive part with a slight permanent strain.

In this material, the cracks are initiated at the macropores and run through the longitudinal yarns and also through the transverse yarns. Because of the wear induced reduction of the interfacial shear stress, the strength of the bridging bundles is reduced and the composite can break after a given number of cycles. At high temperatures up to 1000°C under inert atmosphere (argon), the main evolution of the material is a release of the thermal residual stresses. The microstructural damage is similar to that developed at room temperature, but the level of the interfacial shear stress decreases due to the release of radial thermal stresses.[5–7]

In the SiC/MAS-L composite, the fibres are placed along two perpendicular orientations. When this material is loaded by a uniaxial tension parallel to one direction of fibres, the cracks in the matrix are perpendicular to the loading orientation. A multicracking occurs in the whole section of the specimens, and the fibres in transverse plies are debonded. Due to the mismatch between the thermal expansion coefficient in the matrix and in the fibres, the longitudinal residual thermal stresses are in tension in the matrix, and therefore the transverse cracks are open. During cyclic fatigue, the evolution of the macroscopic mechanical behaviour is controlled by a progressive wear of fibre/matrix interface.[7]

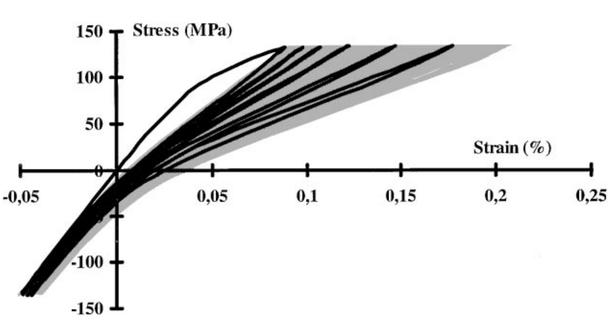

Fig. 2. Experimental evolution of stress/strain loops under tension/compression cyclic loading at room temperature of a pristine SiC/SiC composite (run out after 500 000 cycles, ± 130 MPa, 1 Hz).

In the C/C composite, cracks are mainly initiated around the yarns. In this material, the macroscopic mechanical behaviour is not controlled by the fibres, but by the yarns. Concerning the cyclic fatigue effect, the evolution of the macroscopic mechanical behaviour with the number of cycles applied and the dependence of the life time on the maximum stress applied are explained by a progressive wear of yarn/yarn interfaces instead of the fibre/matrix interfaces. Inside the transverse yarns (±60°) some cracks are also present due to the mismatch between the thermal coefficients of fibre and of matrix. In this material the residual thermal stresses are in slight tension in transverse yarns, hence the cracks inside the transverse yarns are small, and some fibres are debonded.

For the C/SiC composite the structure of reinforcement is similar to the structure of the SiC/SiC composite. But the thermal residual stresses are high in tension inside the tows. This leads to the creation of multiple cracks in the transverse tows after cooling at the end of processing. When this material is subjected to monotonic loading, the main damage created in the microstructure is an extension of the pre-existing cracks, plus a debonding between the carbon-yarns and their surrounding SiC matrix all along the yarns. This debonding occurs as well in longitudinal yarns as in transverse yarns. During a cyclic fatigue test, the same damage is created during the first loading, then a progressive wear of yarn/shell interfaces occurs during the following cycles.

At high temperature (up to 1000°C) under inert atmosphere, the mechanical behaviour of the C/SiC composite under repeated tension is characterized by a strong increase of the residual strain and also by an increase of the mean elastic modulus (see Fig. 3). This evolution depends on the number of cycles and does not depend on the frequency of the cycles. This apparent stiffening induced by cyclic fatigue is generally not observed on materials, but has been observed on the C/SiC composite under repeated tension at 600°C (for stress amplitudes of 110 and 220 MPa), 800°C (for amplitudes of 110 and 220 MPa) and 1000°C (only for an amplitude of 110 MPa) (see Fig. 4), but not at room temperature (for amplitudes of 110 and 220 MPa), at 1500°C (for amplitudes of 110 and 220 MPa) and at 1000°C (for an amplitude of 220 MPa). In order to identify the origin of this stiffening, cyclic fatigue tests have been made under tension/compression (see Fig. 5). In that case, we

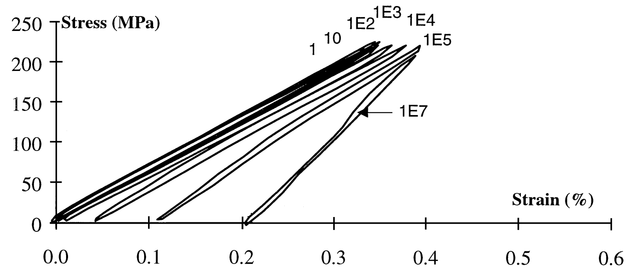

Fig. 3. Experimental evolution of stress/strain loops under tension/tension cyclic loading at 600°C under inert atmosphere (argon) of a pristine C/SiC composite (run out after 10^6 cycles, 0/110 MPa, 1 Hz).

observed that at the beginning of the cyclic fatigue test, the mean elastic modulus measured under compression is higher than the mean elastic modulus measured under tension. Then, during the cyclic fatigue test, the mean elastic modulus under compression remains roughly constant, and during the stiffening stage the mean elastic modulus under tension reaches the value of the mean elastic modulus under compression.

The apparent stiffening is not specific to the C/SiC composite, but has been observed on several materials. This effect has been observed mainly on the C/SiC material at medium temperature, but also on the C/C material at room temperature (see Figs 6 and 7), and on the SiC/MAS-L at 1000°C under inert atmosphere (see Fig. 8). All these materials are characterized by residual thermal stresses in tension in the matrix. On the other hand, this phenomenon has not been observed on the pristine SiC/SiC material, which is characterized by residual thermal stresses in compression in the matrix (see Figs 2 and 10).

In addition, when the SiC/SiC material is heat treated before cycling (for example: 50 h at 800°C under 10^{-2} mbar), the fibre/matrix interface is weakened by a slight oxidation of the fibre coating. This leads to very different behaviour, as shown on Fig. 9. The loop width increases by a certain amount and then decreases.[7] At the same time the residual strain increases, and the slope of the tensile loop increases and reaches a value not far from the compressive slope (see Fig. 10). This macroscopic mechanical behaviour is similar to that observed on the C/SiC at medium temperature (from 600 to 1000°C).

This non-classical stiffening cannot be explained by an interfacial effect in the longitudinal yarns because of the evolution of the mechanical hysteresis during cyclic fatigue. This evolution indicates that the interfacial shear stress between yarn and surrounding matrix (for C/C and C/SiC) or between fibre and matrix (for SiC/SiC and SiC/MAS-L) decreases during the mechanical loading due to wear effects.[5,7] The micromechanical calculations based on a shear-lag model indicate that this interfacial effect leads to a decrease of the mean elastic modulus in tension of longitudinal yarns as observed in pristine SiC/SiC material at room temperature. Therefore the apparent stiffening is difficult to explain by interfacial evolutions between fibre and matrix inside the longitudinal yarns.

On the other hand the analysis of the damage in transverse yarns can explain this phenomenon. In C/SiC, for example, the apparent stiffening is very important. For this material, the transverse yarns contain many cracks after processing. Hence, during a loading/unloading sequence these cracks are opened then closed [see Fig. 11(b)]. By *in situ* trac-

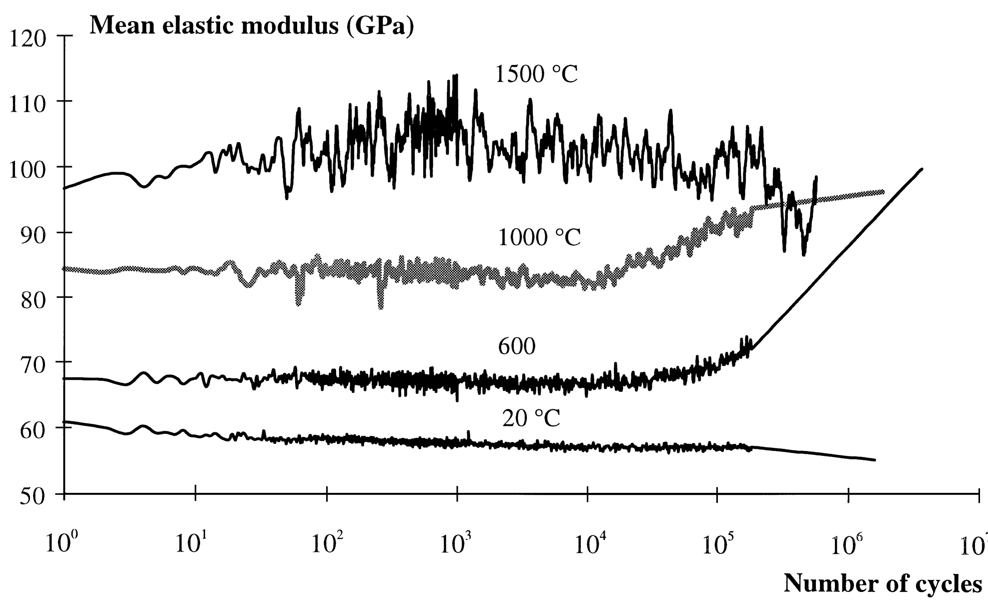

Fig. 4. Experimental evolution of the tensile mean elastic modulus of a pristine C/SiC during tension/tension cyclic fatigue test at constant amplitude ($R=0$) at various temperatures under inert atmosphere (run out after 10^6 cycles, 0/110 MPa, 1 Hz).

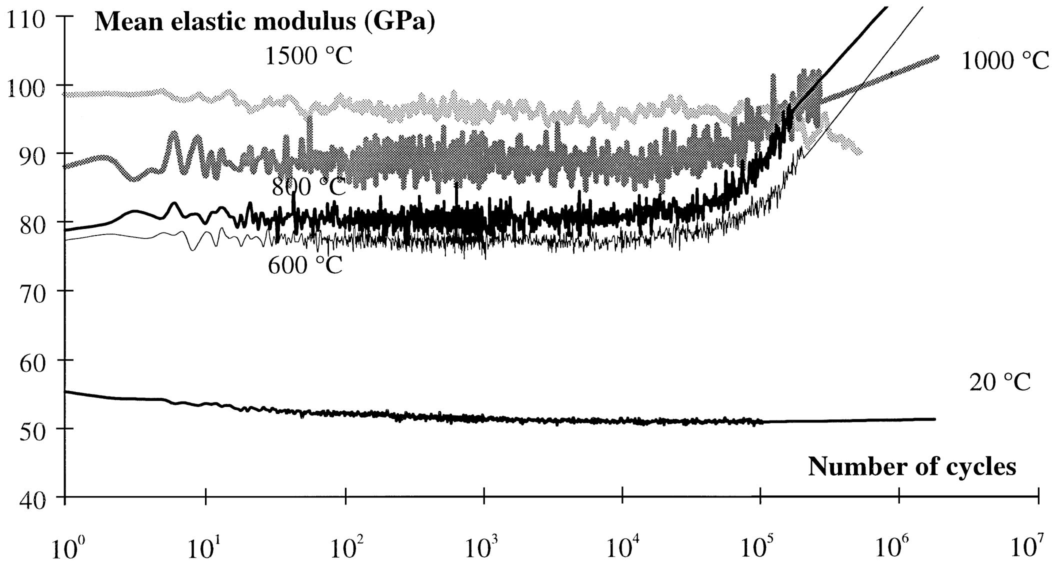

Fig. 5. Experimental evolution of the tensile mean elastic modulus of a pristine C/SiC during tension/compression cyclic fatigue at constant amplitude ($R = -0{\cdot}18$) at various temperatures under inert atmosphere (run out after 10^6 cycles, 0/110 MPa, 1 Hz).

tion under S.E.M.,[8] it has been observed that some fibres in transverse yarns of the C/SiC composite can turn non-reversibly during a loading/unloading sequence. Therefore, during a loading/unloading sequence non-reversible mechanisms operate, and interactions between the lips of a crack in transverse yarns can occur. Moreover, the main effect of medium temperature for the four materials of this study is a release of residual thermal stresses. Hence the opening of the cracks in transverse yarns is lower at high temperature than at low temperature. Instead of an interfacial effect in longitudinal yarns, the stiffening and the high evolution of the residual strain observed during cyclic fatigue at medium temperatures (up to 1000°C) on C/C, SiC/MAS-L and C/SiC composites, can be explained by an increasingly poor closure of cracks in transverse yarns during a loading/unloading sequence [see Fig. 11(c)].

In addition, for the C/SiC, at room temperature the cracks in transverse tows are open due to the thermal residual stresses which are in tension inside the tows. These cracks are open enough to avoid the interactions of their lips. At high temperatures (higher than 1000°C) time-dependent phenomena occur and, due to these specific phenomena, the cracks are also open enough in transverse tows. Then, due to these two different mechanisms, for these last cases the apparent stiffening is not experimentally observed on the C/SiC composite (see Figs 4 and 5).

In order to explain the different experimental results observed on SiC/SiC, SiC/MAS-L, C/C and C/SiC at various temperatures from room temperature to 1000°C under inert atmosphere, a general mechanism can be proposed.

According to the sign of the thermal residual stresses inside the yarns, some cracks may appear in transverse yarns (case of C/C, SiC/MAS-L, C/SiC). If such cracks are present, the stiffening and the large amount of developed residual strain can be explained by an increasing effect of poor closure of cracks in transverse yarns.

Effectively, when the cracks do not exist in transverse yarns, the mechanical behaviour is

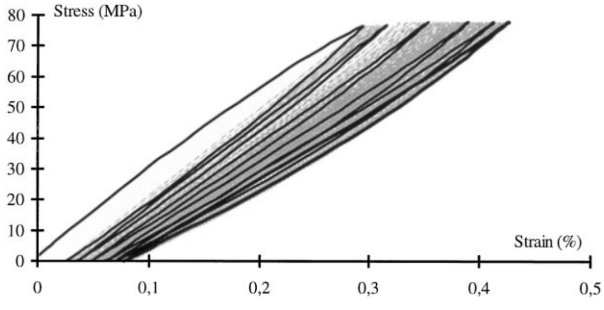

Fig. 6. Experimental evolution of stress/strain loops under tension/tension cyclic loading at room temperature of a pristine C/C composite (run out after $2{\cdot}5 \times 10^5$ cycles, 0/80 MPa, 1 Hz).

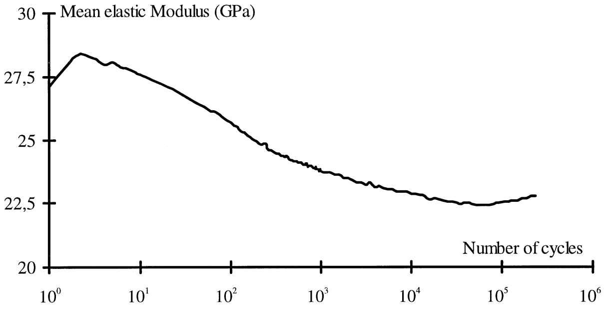

Fig. 7. Experimental evolution of mean elastic modulus under tension/tension cyclic loading at room temperature of a pristine C/C composite (run out after $2{\cdot}5 \times 10^5$ cycles, 0/80 MPa, 1 Hz).

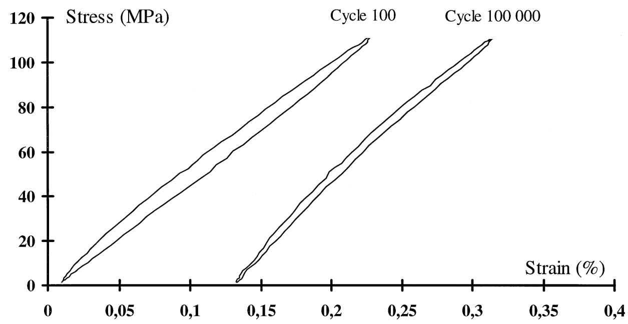

Fig. 8. Experimental evolution of stress/strain loops under tension/tension cyclic loading at high temperature (1000°C) under inert atmosphere of a cross-ply SiC/MAS-L composite (run out after 250 000 cycles, 0/110 MPa, 1 Hz).

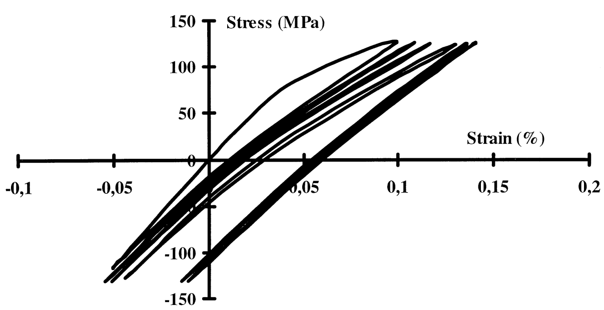

Fig. 9. Experimental evolution of stress/strain loops under tension/compression cyclic loading at room temperature of a SiC/SiC composite after a previous ageing of 50 h under vacuum at 800°C (run out after 500 000 cycles, ± 130 MPa, 1 Hz).

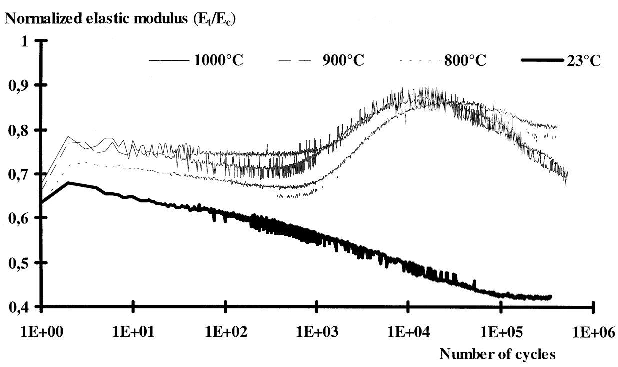

Fig. 10. Experimental evolutions of the ratio [tensile mean elastic modulus (E_t)/compressive mean elastic modulus (E_c)] during cyclic fatigue at constant amplitude of a SiC/SiC previously aged 50 h at various temperatures under vacuum.

completely elastic as well under tensile stresses as under compressive stresses [see Fig. 11(a)]. If some small cracks exist in transverse yarns, these cracks are open under tension and closed under compression [see Fig. 11(b)]. Then the stiffness of the material is higher under compression than under tension. At last, if the cracks inside the transverse yarns are numerous, some irreversible rotations or translations may occur during a loading step [see Fig. 11(c)]. In that case, some interactions occur between the lips of the cracks. The cracks remain open during the unloading step, even under tensile stresses, and the macroscopic mechanical behaviour of the transverse yarns is elastic with the same modulus under tension as under compression. In addition, poor closure of cracks in transverse yarns leads to important residual displacements, that are consistent with experimental results (see Figs 3 and 9).

At the present time this mechanism is only proposed, and should be confirmed by more micromechanical experiments (tension *in situ* under S.E.M., for example). If this mechanism is correct the apparent stiffening should not be observed on unidirectional composites, and this is confirmed by experiments.[9]

5 Conclusions

Usually when a material is subjected to cyclic loading, its mean tension elastic modulus decreases as far as the cyclic fatigue test runs out, in relation to the damage created in the material by the mechanical loading. That is the reason why the mean elastic modulus is commonly used to quantify the damage created in a material. But with certain conditions fulfilled (complex weavy architecture, low interfacial sliding stress) an apparent stiffening can be observed during cyclic loading. This phenomenon is always accompanied by a large amount of inelastic residual strain at zero load. This apparent stiffening is badly understood at present time, but seems to be due to the interlocking of the matrix crack surfaces, giving to the damaged material an apparent elastic behaviour.

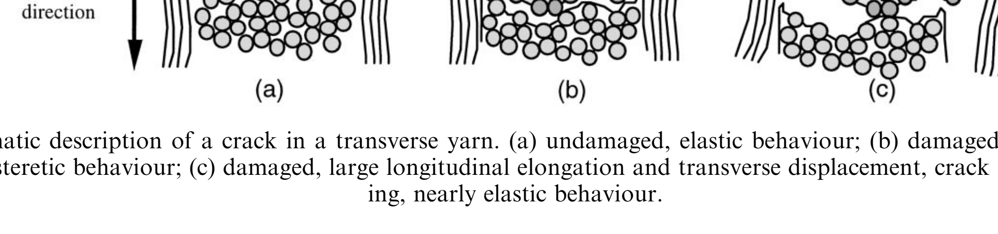

Fig. 11. Schematic description of a crack in a transverse yarn. (a) undamaged, elastic behaviour; (b) damaged, low longitudinal elongation, hysteretic behaviour; (c) damaged, large longitudinal elongation and transverse displacement, crack surfaces interlocking, nearly elastic behaviour.

Acknowledgements

This work has been carried out with the support of the Centre National de la Recherche Scientifique, the Ministère de l'Enseignement Supérieur et de la Recherche, the Direction des Recherches et Etudes Techniques, the Centre National d'Etudes Spatiales, and the companies: Aérospatiale and Société Européenne de Propulsion, within the scientific association 'Thermomechanical behaviour of fibrous ceramic–ceramic composites'. Thanks are particularly due to Dr M. Bourgeon, Dr F. Abbé and Dr J. P. Richard from the Société Européenne de Propulsion, and to Dr P. Pèrés from Aérospatiale, for their interest on this work and for their support.

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