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Thermal fatigue of fibrous ceramic matrix composites

M.L. BOUCHETOU, M. HUGER and C. GAULT

Laboratoire Matériaux Céramiques et Traitements de Surface, URA 320 du CNRS, Ecole Nationale Supérieure de Céramique Industrielle, 47 à 73 Avenue Albert Thomas, 87065 Limoges, France

ABSTRACT

This paper is devoted to the study of 2D SiC/C/SiC composites which are subjected to thermal cycling. Previous work has already shown that variation in Young's modulus can be used to monitor damage in ceramic-ceramic composites. Samples were rapidly heated by moving them up into a tubular furnace and rapidly cooled by moving them down through a gas flow, which can be either air or Ar-H₂. After each thermal cycle, the uniaxial Young's modulus was measured by an ultrasonic method. The results show that a regular decrease of Young's modulus is observed in air up to 100 cycles, though it remains constant in Ar-H₂. In comparison to isothermal ageing, the effects of thermal cycling on oxidation mechanisms are discussed.

INTRODUCTION

A great deal of effort has been devoted in the last 15 years to investigate new materials available for aerospace applications subjected to high stress, high temperature, thermal gradient or severe environmental conditions with especially oxidizing atmospheres. At the present time several fibrous thermostructural composite materials are available ; despite the oxidation phenomena which can affect their mechanical properties. The most promising of them are made with a carbon or silicon carbide matrix. This paper concerns SiC/SiC bidirectional materials⁽¹⁾ in which the reinforcement is ex-PCS SiC fibres⁽²⁾.

The study of the behaviour in service conditions of such high cost materials can be very expensive if it is only based on destructive mechanical tests. Therefore the use of non destructive techniques for characterization of damage presents significant advantages. For measurement of Young's modulus at high temperature, a technique using propagation of ultrasonic waves has been previously developed^[1]. This is particularly suitable for following microstructural evolution in heterogeneous materials, such as composites, during thermal treatments. In the case of the SiC/SiC materials, previous work has shown that the Young's modulus could be significantly affected by the oxidation mechanisms which can occur at the fibre matrix interface during isothermal ageing^[2]. This paper deals with the adaptation of such a technique to the study of SiC/SiC composites during thermal fatigue experiments. In particular, the influence of oxygen on damage is studied.

¹ specially processed for this work by the Société Européenne de Propulsion

² Nicalon, Ceramic grade NLM 202, from Nippon Carbon

EXPERIMENTAL

The bidirectional material is processed in shape of plates. SiC yarns of about 500 fibres were woven at 90°, and then the weaves were stacked before matrix processing. In order to obtain good mechanical properties, fibres were coated with a pyrolytic carbon layer of about 0.1 µm thickness^[3] before preparation of the SiC matrix by chemical vapor infiltration (C.V.I.)^[4] which leads to a SiC/C/SiC composite. For thermal fatigue experiments, samples were machined out of the as-processed plates by cutting with a diamond tool along one of the reinforcement directions.

The measurement of Young's modulus is based on the propagation of ultrasonic longitudinal waves in a "long bar" mode which means that the cross-sectional dimension of the sample must be smaller than the wavelength^[1]. Ultrasonic low frequency pulses are generated by a magnetostrictive transducer welded to a tungsten waveguide on which the sample is glued with a refractory alumina cement (cf. Fig. 1).

Fig.1 : Principle of the ultrasonic line and the associated pattern

In order to preserve the bonding between the waveguide and the sample, the sample has been divided in two parts by machining. The first part acts only as a waveguide and the second part is the effective portion of the sample which is subjected to thermal cycles. To evaluate the uniaxial Young's modulus E , which can be calculated from the ultrasonic velocity V , the time τ between two echoes corresponding to the ultrasonic propagation in the effective portion is measured by a cross-correlation method^[5].

$$E = \rho V^2 = \rho \left(\frac{2L}{\tau} \right)^2 \quad (1)$$

where L is the effective portion length and ρ the density.

In order to impose a thermal flow perpendicular to the weaving plane of the composite, the sample is heated on its "hot face" and is cooled on its "cold face" by water circulation in the metal support (cf. Fig. 2). In a first step, both support and sample are moved up into a tubular furnace (cf. fig. 3) to heat the "hot face". When the temperature of this face, evaluated by thermocouples placed in lateral standard plates (cf. Fig. 4), reaches a chosen temperature $T_{\max}$, both support and sample are moved down in front of a gas blower for cooling. The gas can be air for experiments in oxidizing atmosphere or Ar-H₂ as non oxidizing atmosphere. The cooling is stopped as soon as the temperature of the "hot face" reaches a chosen temperature $T_{\min}$ and then a new thermal cycle can be restarted. A typical temperature cycle is shown on Fig. 5. Young's modulus E is evaluated at the highest and at the lowest temperatures of each

cycle. Then the relative variation of E (normalized to the room temperature value E_0) is plotted versus the cycle number N . In this paper, only relative variations of E evaluated at T_{min} are reported.

Fig. 2 : Water cooling system

Fig. 3 : Thermal fatigue equipment

Fig. 4 : Cross section of the system

Fig. 5 : Typical temperature cycle

RESULTS AND DISCUSSION

Thermal fatigue experiments have been performed in air with different values of T_{max} : 750°C, 800°C, 900°C, and 1000°C (cf fig.6). During the first few cycles, the relative variations of Young's modulus are rather small. E then remains constant up to 100 cycles for $T_{max}=750^\circ\text{C}$, while a linear decrease occurs when T_{max} is above 800°C.

Fig.6 : Young's modulus relative variations during thermal fatigue in air for different chosen temperatures T_{max}

Ultrasonic measurements of Young's modulus have been performed previously on this material during isothermal ageing at different temperatures under oxidizing or neutral atmosphere^[2]. Those experiments showed the importance of the environment on the behaviour at high temperature. For ageing below 1200°C , Young's modulus remains constant in a non oxidizing atmosphere, while significant changes in Young's modulus occur in oxidizing atmosphere. Usually, relative variations of Young's modulus versus time (cf. Fig.7) exhibit three stages which have been interpreted as follows in agreement with results obtained by other authors ^[6, 7]:

Fig.7 : Young's modulus relative variations during isothermal ageing in air^[8]

In the case of isothermal ageing in air below 1200°C , it has been established that the cause of these Young's modulus variations is the oxidation phenomenon at the fibre matrix interface^[8]. Furthermore, the constant value of E observed in a non oxidizing atmosphere confirms that no damage occurs during isothermal ageing without oxygen in this temperature range. But in the case of thermal fatigue, the thermal

gradient applied to the sample can generate microcracks in the matrix which usually lead to significant decrease of Young's modulus, according to the formula derived by Hasselman^[9] :

$$E = E_i \cdot \left(\frac{1}{1 + a \cdot \mu \cdot \langle r \rangle^3} \right) \quad (2)$$

where E_i is the Young's modulus of the crack free material, a and r are constants depending on geometry and orientation of the microcracks, and μ is the microcrack density.

In order to separate the damage due to thermal fatigue from the damage due to interphase oxidation, experiments in a non oxidizing atmosphere (Ar-H_2) have been performed for the two highest $T_{\max}$ values (900°C and 1000°C). As shown in Fig.8 and Fig.9, Young's modulus remains constant in Ar-H_2 up to 100 cycles for the two considered temperatures, though a regular decrease of E is observed in air.

Fig.8 : Young's modulus relative variations during thermal fatigue for a chosen temperature $T_{\max} = 900^\circ\text{C}$

Fig.9 : Young's modulus relative variations during thermal fatigue for a chosen temperature $T_{\max} = 1000^\circ\text{C}$

These results confirm that the pyrocarbon interphase oxidation is the main phenomenon which is responsible for the damage observed during thermal cycling. The slight decrease of Young's modulus which often occurs during the first few cycles (cf. Fig.6) could be attributed to the formation of microcracks in the matrix. However the constant value of E observed in Ar-H_2 shows that this damage which could be generated by the thermal gradient inside the sample remains very low. Nevertheless it could play an important role on the subsequent decrease of Young's modulus in air by opening new channels for oxygen access which enhance pyrocarbon interphase oxidation.

CONCLUSION

Since many potential applications of SiC/C/SiC composites involve repetitive thermal cycles, experiments have been performed in order to investigate the damage which can be generated by such a treatment. Young's modulus is a good indicator to follow the evolution of damage during thermal cycling. Results show that very little damage occurs when experiments are conducted in non oxidizing atmosphere, but an important decrease of Young's modulus is observed in air. By comparison with previous results obtained during isothermal ageing in a similar atmosphere, the pyrocarbon interphase oxidation is undoubtedly the main phenomenon responsible for this damage. Nevertheless it seems that the oxidation rate can be increased by the thermal fatigue. Indeed, even if there are only a few microcracks, these can have a strong influence on the oxidation rate by opening new channels for oxygen accesses.

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