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High temperature creep performance and microstructure of SiC-C-SiC composites

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ABSTRACT

The high temperature creep behaviour of a 2D SiC/C/SiC composite is studied at temperatures ranging from 1200 to 1673K, under stresses from 50 to 500MPa. Due to the microstructural evolution of the material, steady state creep is never reached; creep toughening is observed. Microcrack formation and propagation in the interface and in the matrix were identified as contributing to the deformation.

1. INTRODUCTION

The range of applications where ceramic matrix composites are used has considerably grown over the last twenty years, specially in the automotive, aeronautic and aerospace industries. The first generation of these composites, containing continuous carbon fibre reinforcement, was too sensitive to oxidation and was eventually replaced by more oxidation-resistant composites for many applications. The carbon matrix was replaced by a SiC matrix (CVD or CVI) and the carbon fibres by SiC fibres, a thin interfacial carbon layer being added to allow easy debonding of the interface. Very high fracture strength, as well as decent resistance to oxidation, were conferred to this class of composites by carefully engineering the microstructure of the material: control of the cristallinity and composition of the fibre, of the thickness and structure of the carbon interphase and of the porosity of the matrix. For instance, special geometries, like the interwoven two-dimensional network of Nicalon fibres, were designed. Since application of these composites as structural materials are considered at temperatures up to 1600K, with a lifetime of hundreds of hours, not only are the high temperature corrosion of these materials by the surrounding atmosphere and their fracture resistance important properties, but as well their high temperature creep resistance.

It is precisely the object of this work to study the high temperature creep behaviour of a 2D Nicalon-fibre/C/SiC(CVI) composite, to understand the damage mechanisms involved and to predict long time behaviour.

2. EXPERIMENTAL DETAILS

2.1. Material

The 2D SiC-C-SiC composite is provided by SEP, France, as 3mm thick plates. It is made of stacks of two dimensional nets, made of NLM 202 Nicalon fibre bundles. This fabric-like nets are coated by

chemical vapour infiltration with a carbon layer about $0.1\mu\text{m}$ thick. The SiC matrix is afterwards deposited in three subsequent cycles by chemical vapour infiltration to reach a final density of $2.5\text{--}2.6\text{g/cm}^3$. The material porosity is on the order of 10%. Its total fibre content is 40vol%. A typical structure of the composite is shown in Fig. 1.

Fig. 1: In plane and cross sectional view of the 2D SiC/C/SiC composite

2.2. Creep experiments

Compressive creep tests are performed at three temperatures: 1123, 1423 and 1673K, at stresses between 50 and 500MPa and under argon flow. The $2\text{mm}\times 2\text{mm}\times 5\text{mm}$ samples with planparallel sides are deformed under constant load in compressive mode with the fibre net (yz plane) either parallel or perpendicular to the deformation axis. Since the composite microstructure evolves with the annealing time, each sample is deformed for fixed conditions of load and temperature.

The constant-load creep apparatus used for this investigation is described in detail in (1). It comprises a furnace with lanthanum chromite heating elements. The load is applied onto the sample via silicon carbide push rods, two silicon carbide platelets ($10\text{mm}\times 10\text{mm}\times 4\text{mm}$) being placed between the push rod and the sample end-faces. The entire set-up is enclosed in a gas-tight alumina tube located inside the furnace, so that the atmosphere around the sample can be controlled. All tests are run under continuous argon flow. A thermocouple located near the sample is used to control the furnace temperature. The shortening of the sample is recorded by a direct current deformation transducer. At the end of each experiment, the total recorded deformation is compared to the final length of the sample.

2.3. Sample observation

The starting material and the differently annealed and deformed materials are investigated by scanning electron microscopy (SEM) and, for special conditions, by transmission electron microscopy (TEM). SEM observations are always made in the three different planes of the composite, xy, xz and yz, which are indicated in Fig.2.

3. EXPERIMENTAL RESULTS AND DISCUSSION

3.1. Microstructure of the starting material

The Nicalon fibres are assembled in torons of about 500 filaments, which are woven to the two dimensional network. The longitudinal torons can thereby be either in phase or out of phase, producing by this irregular architecture a distribution of large pores. The fibre diameters vary from 10 to $26\mu\text{m}$. Electron diffraction and high resolution imaging in the TEM indicate that the fibre is nanocrystalline, consisting of small silicon carbide crystals (average size 3nm) and even smaller carbon crystals (1nm or less) embedded in an amorphous oxycarbide phase. The entire fibre is surrounded by a thin carbon layer

of $0.1\mu\text{m}$. The carbon is of turbostratic character. The SiC CVI matrix consists of small equiaxial grains next to the fibre surface, surrounded by large columnar grains (2-3-4-5).

Fig. 2: Schematic presentation of the different observation planes in the composite

3.2. Microstructure of the annealed material

Upon annealing in the creep apparatus (under argon flow without application of a load) at the different temperatures, the microstructure of the composite evolves. Starting from the outer surface, the interfacial carbon layer reacts and forms of a silica layer on the fibre side and a layer of large SiC crystals on the matrix side. As a result of this reaction, a free gas space is left between the two layers, which allows fast displacement of the reaction front (at 1673K, the reaction develops over a depth of 1mm in 50h). In addition, the size of the SiC nanocrystals in the fibers increases by Ostwald ripening (at 1673K, they reach 50nm in 24 hours) and the global oxygen content of the fibre slowly decreases to a level in equilibrium with the surrounding atmosphere (6).

3.3. Creep results

At all investigated temperatures, the creep curve shows three typical stages (Fig.3): a primary stage where the creep rate decreases with time, a pseudo steady-state stage characterized by a very small variation of the creep rate, and finally a tertiary stage corresponding to sample fracture. At 1673K, sample fracture occurs at 300MPa when the fabric-like layer is parallel to the deformation axis and as early as 100MPa if this layer is perpendicular to the deformation axis. Real steady-state creep (constant creep rate) was never observed. The creep rate always slightly decreases, corresponding to a hardening which must be related to the microstructural evolution of the material at high temperature, see 3.2. Fig.4 shows $\log \dot{\epsilon}$ curves at 1673K, 1423K and 1123K, at different times of the quasisteady-state range, indicating the hardening effect. The same hardening effect is obtained when the sample undergoes ageing prior to the creep deformation. Even though a power law behaviour is not strictly followed we will describe the creep behaviour of the composite by this approach: At high temperature, in the low stress range, the stress exponent is very low, about 0.5, while in the high stress range it is almost 2. At lower temperature, the stress exponent remains small.

3.4. Microstructure of the deformed material

1673K, $\sigma // y$ or z : At 1673K and low stress, on the order of 100MPa, in addition to decohesion of the fibre interface (due to the annealing), few cracks were observed in the matrix in xz and yz sections which interconnect the fibres in torons perpendicular to the applied stress, showing an efficient crack deviation in the fibre interfaces. Only very few fibres are fractured. With increasing stress, a multiplication of the matrix cracks in the yz and xz planes parallel to the applied stress is observed with a frequent bifurcation at the fibre interfaces. In addition, matrix cracks start to occur as well in the xy plane. They become

increasing stress, fibre fracture becomes more and more important, yielding more and more possible fracture surfaces in the fibre and a segmentation of the fibres by these cracks. At about 200MPa, cracks along the limits of the matrix infiltration cycles were observed and some large cracks parallel to the deformation axis which propagate all the sample. The crack density in the xy plane is still lower. At even higher stresses, like 300MPa, the cracks parallel to the stress axis propagate directly through the matrix and through the fibre, without any bifurcation or deviation in the fibre interface. Failure is finally obtained by lamination of the torons para/perpen to the applied stress. Typical micrographs of a sample deformed at 200MPa are shown in Fig.5. Preannealing of the samples shifts the above described behaviour to lower stresses, for example at 1673K, complete failure is already observed at 200MPa.

Fig.3: Presentation of the composite deformation as a function of time at 1223K, 386MPa, fibre net parallel to the deformation axis

Fig.4: Log-log presentation of the creep rate as a function of stress for different temperatures and the fibre net either parallel or perpendicular to the creep axis

4. CONCLUSION

Extended microstructural analysis of the as-received, annealed and under different temperature and stress conditions deformed composite have allowed to determine the main mechanisms which contribute to the high temperature compressive creep of the 2D SiC/C/SiC composite.

Decohesion of the fibre interfaces occurs already during high temperature annealing as a result of the chemical reactions which lead to the consumption of the carbon interphase and its substitution by a layer of SiC crystals embedded in an amorphous silica matrix. This decohesion and special the interfacial crack opening is even accentuated under the influence of an applied stress.

Already at low stresses, small microcracks form and propagate in the CVI SiC matrix perpendicular to the fibres, interconnecting the fibres and propagation in the fibre interfaces (fibre debonding). Bifurcation of the cracks may occur at the fibre interfaces. The crack multiplication increases with increasing stress. With increasing stress, cracks start to propagate along the border of the infiltration cycles, yielding wide interconnected networks of matrix cracks.

This low and intermediate stress domain corresponds to a region in the creep rate stress diagram with a stress exponent of only 0.5.

In the high stress domain the creep exponent becomes 1.8. The creep rates correspond to those of other composites with similar fibres. In samples which were deformed in this domain more and fibre damage is observed, going up to the final fracture of the fibres themselves. Therefore, we conclude, that at high stresses fibre creep and final fracture of the fibres determine the composite behaviour.

XZ plane

XY plane

YZ plane

Fig.5: Microstructure of the composite deformed at 1673K, 200MPa with the fibre net parallel to the deformation axis, views of the XY, XZ and YZ planes

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