Available online at www.sciencedirect.com
ScienceDirect
Journal of Nuclear Materials 367–370 (2007) 672–676
journal of nuclear materials
www.elsevier.com/locate/jnucmat

Characterization of commercial grade Tyranno SA/CVI-SiC composites

B. Riccardi [a,*], E. Trentini [b], M. Labanti [b], M. Leuchs [c], S. Roccella [a], E. Visca [a]

[a] Associazione EURATOM-ENEA, P.O. Box 65, I-00044 Frascati (RM), Italy
[b] ENEA C.R. Faenza, I-48018 Faenza (RA), Italy
[c] MT-Aerospace AG, Augsburg, Germany

Abstract

The objective of the present work was to characterize commercial-grade Tyranno SA SiC fiber reinforced chemically vapour infiltrated (CVI) SiC matrix composites (SiC_f/SiC) with chemically vapour deposited (CVD) SiC coating. The characterization includes the assessment of the monotonic mechanical properties. Low cycle flexural fatigue behaviour has been investigated at room temperature (RT) and 1000 °C by means of 4-point bending tests. The creep behaviour at 1000 °C was preliminary investigated by means of constant bending stress rupture test. The material showed a pronounced degradation of monotonic mechanical properties at high temperature. Low cycle flexural fatigue behaviour showed excellent and satisfactory results at RT and 1000 °C, respectively. The creep resistance at 1000 °C is significant only at low load level.

1. Introduction

SiC_f/SiC composites are one of the most promising candidates for fusion structural materials because of their potential application for high performance reactors and good safety characteristics compared to metallic materials. Favourable features of SiC_f/SiC composites are the high temperature properties and the low activation characteristics at short and medium term [1]. Conversely, the material has some critical issues such as the high gas production due to nuclear transmutation and properties degradation upon neutron irradiation. Continuous progress in R&D and, in particular, the availability of advanced fibers and improved processing methods has achieved higher thermo-mechanical characteristics and better irradiation stability of the composites.

Since the beginning of the effort on SiC_f/SiC composites in the European fusion technology program, the focus has been on the development of commercial grade composites for fusion reactor applications [2]. Following the R&D on ceramic grade SiC fiber composites, the development of a new EU reference material started when the new stoichiometric SiC fibers become widely available in the industry. In particular, the effort was focused on the development of composites employing 2D and 3D fabrics of UBE Grade 3 Tyranno SA fibers (UBE Industries, Japan). The activity was a joint venture between FN Spa (Italy) and MAN Technologie AG (now MT Aerospace AG, Germany) at the

* Corresponding author. Tel.: +39 06 94005159; fax: +39 06 94005147.
E-mail address: riccardi@frascati.enea.it (B. Riccardi).

0022-3115/$ - see front matter © 2007 Elsevier B.V. All rights reserved.
doi:10.1016/j.jnucmat.2007.03.098

end of 2002 and was carried out during the following two years. The new material was finally delivered and is being characterized extensively by several EU Fusion Associations; this characterization concerns thermo-mechanical testing including creep and fatigue and electrical testing of unirradiated materials. The material is also undergoing neutron irradiation at high temperature and fluence in OSIRIS and Phenix reactors at CEA (France).

2. Experimental

Two dimensional (2D) performs were obtained by stacking and pressing plain weave cloths made of Tyranno SA-3 fibers up to the fiber volume fraction of about 40% and a thickness of 3–4 mm, respectively. Three dimensional (3D) preforms were woven by Shikibo (Japan) and were characterized by about 35% fiber volumetric fraction and about 10% of the total fiber amount through the preform thickness.

All the preforms were subjected to a short isothermal CVI run to form a thin (about 80 nm) carbon interphase followed by isothermal CVI for matrix densification. Finally a SiC coating of about 80 μm thickness was over-coated by CVD. The final mean thicknesses of the 2D and 3D composites were 3.7 and 4.5 mm, respectively. The apparent density was 2.70 g/cm^3 for the 2D composite and 2.65 g/cm^3 for the 3D; the total porosity was about 13% for the 2D composite and slightly higher for the 3D.

The CVI process leads to intrinsically porous composites with low thermal diffusivity/conductivity but a previous work has shown that it is possible to improve thermal diffusivity/conductivity by using stoichiometric fibers and 3D textures [3]. When compared with commercial ceramic grade fiber reinforced CVI composites [4] the material manufactured here showed a significant improvement in thermal conductivity both for 2D and 3D composites. In fact, the RT transverse conductivity values estimated from laser flash diffusivity measurements were 15 W/m-K for the 2D and 28 W/m-K for the 3D composite [5].

Preliminary low cycle fatigue testing has been undertaken to investigate the fatigue behaviour of the 2D composites. Four-point flexural cyclic tests have been carried out at room temperature and 1000 °C in pure Ar environment by using a sinusoidal wave form stress amplitude and a frequency of 2 Hz. The test duration was limited to 0.8×10^5 cycles. The peak stress imposed for testing ranged between a significant and a small fraction of the unfatigued flexural strength. A small load (50 N), corresponding to a minimum stress of about 30 MPa, was imposed in order to keep the specimen in place in the bending fixture.

Preliminary stress rupture testing has been undertaken to investigate the creep behaviour of 2D composite. The tests have been carried out by using 4-point bending according to CEN-EU Standards for ceramic materials [6]. The testing temperature was 1000 °C under pure Ar gas. The total test cycle consisted in a heating up to 1000 °C (4 °C/min), 60 min hold time, load application, testing, natural cooling (with flowing Ar gas). The applied bending load ranged from 150 to 60 MPa.

3. Results and discussion

3.1. Monotonic mechanical testing

All composites were preliminarily characterized to investigate the mechanical properties (Table 1). The respective Young's modulus and tensile strength were evaluated at room temperature by using full thickness 100 mm long × 10 mm wide × 3–4 mm thick bars according with DIN 50160 standard. The mean values of Young's modulus and tensile strength were 293 GPa and 272 MPa for the 2D composite and 198 GPa and 252 MPa for the 3D. A typical 2D tensile stress–strain curve is illustrated in Fig. 1 and shows good initial linear behaviour, but small elongation.

Through-thickness tensile strength was assessed by using 20 mm diameter, full thickness discs and gluing the specimens to steel profiles by means of epoxy glue. The values measured for 2D and 3D

Table 1
Mechanical test results

Composite	2D	3D
Fiber volume (%)	41	35
Density (g/cm^3)	2.73	2.65
Young's modulus (GPa)	295	252
Matrix cracking stress (MPa)	110	n.a.
Tensile strength (MPa)	272	198
Interlaminar shear strength (MPa)	54	45
Through thickness strength (MPa)	7,6	16.6
Bending strength at RT (MPa)	356 ± 47	n.a.
Bending strength at 600 °C (MPa)	303 ± 11	n.a.
Bending strength at 1000 °C (MPa)	268 ± 27	n.a.
Strain to failure at RT, 600 °C, 1000 °C (%)	0.19, 0.16, 0.15	n.a.

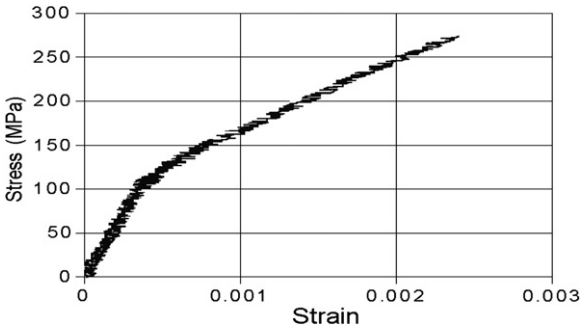

Fig. 1. RT tensile stress–strain curve for 2D composite.

were 7.6 and 16.5 MPa, respectively. Few representative results were recorded because the failure occurred generally in the glue layer at low load. The interlaminar shear strength was measured by three point short span (14.5 mm) bending testing by using 30 mm long × 10 mm wide, full thickness bend bars. The values obtained were 54 and 45 MPa for the 2D and the 3D, respectively.

Four-point bending tests on 2D composites were carried out at RT, 600 °C and 1000 °C according to ASTM C1341-96 test method (inner and outer spans were 20 mm and 40 mm) by using full thickness specimens. The high temperature testing were carried out in flowing pure argon ($H_2O = 3$ wppm, $O_2 = 2$ wppm, hydrocarbons = 5 wppm, Ar = balance). The bending strength results are shown in Table 1, the measured stress and strains were calculated according with ASTM standards considering linear behaviour of the material. The results of tests show a significant decrease of bend strength with increasing temperature. The standard deviations are large, but not related to the testing temperature. The strain to failure is very low at all testing temperature. Scanning electron microscopy (SEM) observations of fracture surface (Fig. 2) show differences with respect to the test conditions; although the pull out length appears rather limited in all tests, the specimens fractured at RT show a slightly larger pull out compared to the ones fractured at 1000 °C. A possible explanation of the observed flexural behaviour can be attributed to the limited thickness of the carbon interphase and the external roughness of Tyranno SA-3 fibers.

3.2. Cyclic tests

Few results are available on fatigue behaviour of unirradiated SiC_f/SiC composites and these results

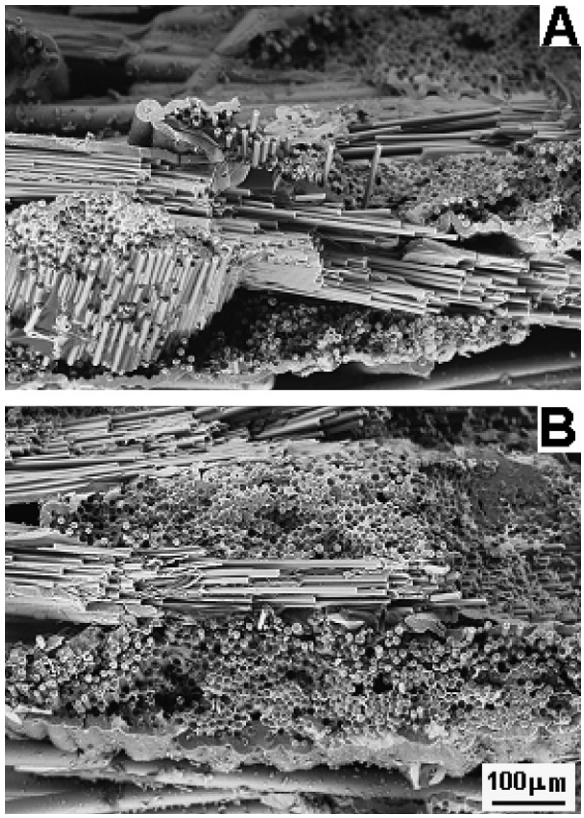

Fig. 2. Fracture surface after monotonic bending testing at (A) RT and (B) 1000 °C.

support the observation that the number of cycles to failure is generally a function of the peak stress and that the fatigue stress threshold is below that at which crack growth does not occur [7]. In our experiment, the results at RT have shown that the material has a good fatigue behaviour, as no failure occurred after 0.8×10^5 cycles at a peak load up to 98% of the flexural strength, a value well above the matrix cracking stress (110 MPa). This result is consistent with an early study [8]. It indicates that the composite can avoid the matrix crack propagation produced in the first loading cycle because the microcracking process is arrested during cyclic loading. The crack can remain stationary because fiber bridging reduces stress intensity at its tip. The stress–strain loops indicate that at each loading and reloading cycle the tangential elastic modulus remains practically constant, showing that the behaviour is dominated by an effective fiber-interface sliding since the loop area remains almost unchanged.

In contrast with the results at RT, when tested at 1000 °C, specimens failed before reaching 0.8 ×

10^5 cycles even at a stress level well below the 1000 °C flexural strength. This different behaviour indicates that the composite suffers from matrix crack propagation; in fact, the microcracking process does not reach a stationary condition during cycling because the fiber bridging force on the cracks is reduced due to the fiber creep. The results obtained at 1000 °C are summarized in Fig. 3 where peak stress versus fatigue life (Wöhler curve) is plotted. A high lifetime is expected only for stress as low as 40% of the 1000 °C flexural strength. The stress–strain loops (Fig. 4) indicate that specimens deteriorate continuously even if they are still able to sustain the fatigue load. The increase of the loop area with increasing fatigue cycles indicates a degradation of the inter-fiber–matrix strength. The failure is commensurate with a decrease of tangential elastic modulus of about 70%. The fracture surface of such specimens indicates that fiber pull out is nearly absent.

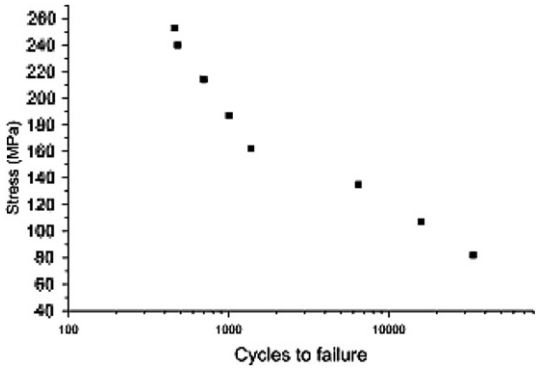

Fig. 3. Stress versus cycles to failure at 1000 °C for 2D composites.

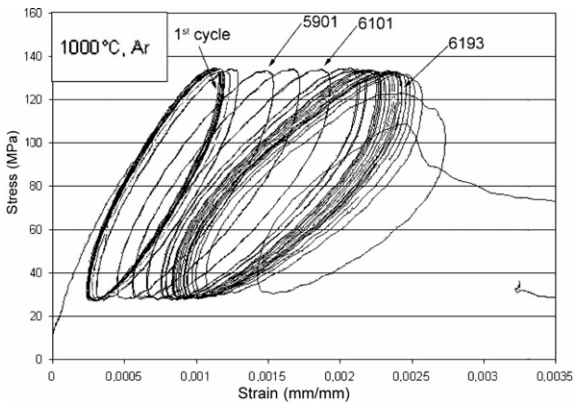

Fig. 4. Stress–strain loops (1000 °C, 50% MOR).

3.3. Flexural stress rupture test

The obtained curves (strain versus time) are shown in Fig. 5. With the exception of the curve at 150 MPa, which shows fast failure, two stages can be observed: the first (primary stage) is related to a significant increase of the strain during the initial part of the test; the second (stationary stage) is related to a stabilization of the strain rate. The lifetime diagram, i.e. the applied stress versus lifetime is shown in Fig. 6. The outcome of the tests is that the material shows a limited stress rupture resistance. In fact, a significant time to rupture can be extrapolated only at stress values well below the matrix cracking stress. The analysis of the fracture surfaces revealed that the failure was almost completely brittle with a nearly no fiber–matrix debonding. At high load, the lifetime is only few hours. In such a condition [9], the stress rupture behaviour

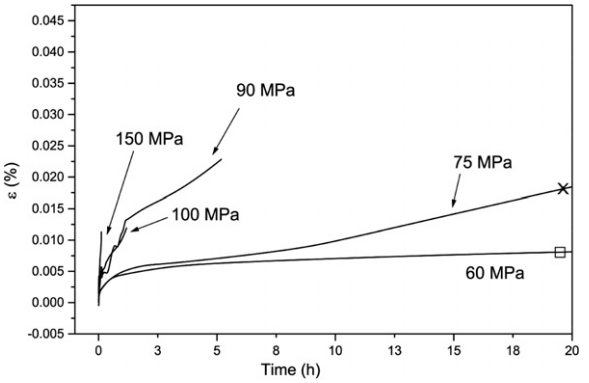

Fig. 5. Strain versus time during stress rupture testing at 1000 °C [(×) failure at 46 h time, (□) on going].

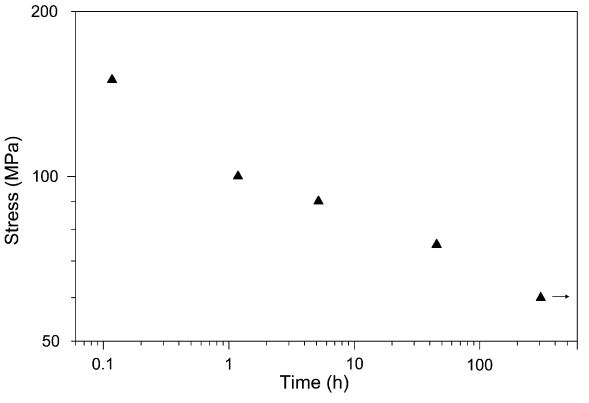

Fig. 6. Stress versus rupture time for 2D composites [(→) on going].

is dominated by an extensive matrix cracking that occurs during the initial application of the load. Since the interfacial shear strength of the composites is very high, due to fiber roughness and interphase thickness, the bridging fiber sliding is modest and therefore fibers start to creep under the load induced by the extensive matrix cracking. Therefore, the fiber creep strain increases and the fiber creep rupture is reached in very short time. Finally, the progressive matrix reloading is high enough to cause full matrix cracking and the specimen failure.

At low load, the lifetime increases significantly (46 h at 75 MPa). In this case, the composite behaviour is determined by a limited matrix cracking produced when the load is applied. During the consequent creep of the fibers bridging the cracks, the time dependent redistribution of stress from fibers to matrix determine the matrix crack opening and the loading of additional undamaged fibers. In these conditions, due to the further fiber creep, the fibers start to break at stress concentration sites. When the fiber rupture reaches a critical level, the heavy reloading of the matrix leads to the final fracture of the specimen.

4. Conclusions

Commercial grade 2D and 3D Tyranno SA fiber/CVI-SiC composites have been manufactured. The mechanical properties satisfy the specification for the material procurement for fusion reactor applications, but the material showed a pronounced degradation of properties at high temperature. Low cycle flexural fatigue behaviour investigated by means of 4-point bending tests showed excellent and satisfactory results at RT and 1000 °C, respectively. The creep behaviour, investigated by means of constant load stress rupture test by 4-points bending tests, showed only a short lifetime at high stress and became significant only at very low load. The material is undergoing extensive characterization but the preliminary results shown in the paper indicate that further R&D on fiber-matrix interphase is necessary to improve the mechanical behaviour.

Acknowledgement

This work, supported by the EURATOM Communities under the contract of Association between EURATOM/ENEA, was carried out within the framework of the European Fusion Development Agreement. The views and opinions expressed herein do not necessarily reflect those of the European Commission.

References

[1] A.R. Raffray, R.H. Jones, G. Aiello, M. Billone, L. Giancarli, H. Golfier, A. Hasegawa, Y. Katoh, A. Kohyama, B. Riccardi, M.S. Tillack, Fusion Eng. Des. 55 (2001) 55.
[2] L. Giancarli, H. Golfier, S. Nishio, R. Raffray, C. Wong, R. Yamada, Fusion Eng. Des. 61&62 (2002) 307.
[3] R. Ramada, N. Igawa, T. Taguchi, J. Nucl. Mater. 329–333 (2004) 497.
[4] J.H.J. Hegeman, P.G. de Heij, D.S. d'Hulst, M. Jong, J.G. van del Laan, M. van Kranenburg, Fusion Eng. Des. 69 (2003) 403.
[5] B. Riccardi, Report ENEA/FUS-FUS-TN-MA-R-RD-002, 2005.
[6] CEN, European Committee for Standardization, ENV-658-3:2003.
[7] R.H. Jones, D. Steiner, H.L. Heinish, G.A. Newsome, H.M. Kerch, J. Nucl. Mater. 245 (1997) 87.
[8] S. Zhu, M. Mizuno, Y. Kagawa, J. Cao, Y. Nagano, H. Kaya, J. Am. Ceram. Soc. 79 (1996) 3065.
[9] S. Zhu, M. Mizuno, Y. Kagawa, J. Cao, Y. Nagano, H. Kaya, Mater. Sci. Eng. A225 (1997) 69.