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Comparison of the mechanical behaviour of SiC_f/SiC composites following neutron irradiation and helium implantation

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Abstract

Thermomechanical behaviour of 2D-SiC_f/SiC composites following neutron irradiation and helium implantation was studied. Three-point bending tests were carried out at room temperature (RT) and 1073 K, after neutron irradiation (1.29, 2.69 and 5.23 dpa) at the High Flux Reactor–Petten. Results of RT mechanical tests on helium implanted bars are also referred and discussed. The samples with an accumulated damage dose of 1.29 dpa show a strength decrease and improved bend toughness. For the higher neutron fluences the composite exhibits a brittle behaviour. For helium implanted materials, a strength decrease comparable to that observed for those neutron irradiated at lower fluence is present, but its toughness is reduced.

1. Introduction

Interest in the low activation of SiC and availability of SiC-based composites with predictable high-temperature properties has motivated the studies of this class of materials for fusion structural applications. Low activation characteristics of current industrial SiC_f/SiC composites produced by Chemical Vapour Infiltration (CVI) already approach those predicted for impurity-free SiC [1,2].

In operating conditions, the material is subject to neutron irradiation, with an energy range as high as 14 MeV. This causes both displacement damage and the production of helium in SiC, via transmutation reactions. In order to assess the effects of direct radiation damage and helium production on the mechanical properties of the structural materials, a testing campaign has been conducted. Post-irradiation mechanical testing (three-point bending) was carried on "as-received" and neutron irradiated specimens with accumulated damage levels of 1.29, 2.69 and 5.23 dpa. The former results are compared with data obtained for SiC/SiC samples, from the same production batch, after fusion relevant helium implantation. Implantation doses around 2500 appm were obtained with an irradiation temperature of 1175 ± 50 K and an accumulated damage dose of 0.4 ± 0.1 dpa due to the helium implantation.

2. Experimental

The composites considered in our study were produced by SEP [2] using a two-dimensional woven laminate of Nicalon [3] fibres in which a β-SiC matrix is deposited by Chemical Vapour Infiltration (CVI). The matrix infiltration process (CVI) leads to a morphology that follows the two-dimensional yarn configuration with the presence of visible inter-bundle macroporosity, a common feature of CVI processed materials. Before matrix infiltration, a carbon coating (~0.1 μm) is deposited

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[2] Cerasep™, Société Europeéne de Propulsion, France.

[3] NL-202, Nippon Carbon, Japan, is a Si–C–O fibre.

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onto the fibres by chemical vapour deposition to improve mechanical behaviour. The material was supplied as bending specimens with nominal dimensions of $60 \times 8 \times 1.2$ mm^3. The SiC_f /SiC-specimens consisted of five stacked layers from plain weave cloths, consisting of an 800 *denier* Nicalon yarn type with ∼220 fibres each. The fibre content of the composite was 28 ± 3% vol.

Neutron irradiation of the SiC_f/SiC bars was carried out at High Flux Reactor, Petten (HFR). Samples were equally distributed in three identical stainless steel holders, loaded in a TRIO capsule [3]. This device offers the possibility of simultaneous irradiation of the three different sample holders whilst allowing individual control for each holder. The capsules were loaded in the HFR core position C3 and according to the sample holder, total fluences of 1.34×10^{25}, 2.71×10^{25} and 5.41×10^{25} n/m^2 were obtained. Corresponding accumulated damage doses are 1.29, 2.69 and 5.23 dpa. The displacement energies considered were those from previous work [4]. An average temperature of 1023 ± 40 K was registered for the specimens during irradiation and kept constant by adjusting the composition of the helium–neon gas mixture flowing in the gaps between the samples and irradiation channels [5]. Moreover, a vertical displacement unit allowed the sample columns to be moved, in order to follow the shift of the neutron fluence rate curve due to reactor operation. Eight thermocouples per sample holder continuously monitored the actual temperature for the SiC_f/SiC composites. Flux monitors, loaded in one holder, allowed comparison between damage estimates and actual reached values. Details on the irradiation history, statistical evaluation of the temperature distribution during the irradiation are given in detail in a separate report [6].

Helium implantation up to doses of 2500 appm was carried out at the IAM-Ispra Cyclotron, at temperatures of 1173 ± 50 K. For each sample, a homogeneous 0.6 mm deep implanted zone was obtained, extending 8 mm in width and 4 mm in length. Accumulated damage due to implantation was 0.4 ± 0.1 dpa, calculated with the TRIM code [7] and using the same displacement energy values used for neutron irradiation [4]. The implanted volume is located along the central length of the bend bars, so that post-irradiation three-point bending test was possible. Details of the helium implantation experiment can be found in separate papers [8,9].

Mechanical behaviour was evaluated using a high temperature 3-point bend test apparatus [10]. All tests were carried out with a cross-head speed of 0.5 mm/min. Neutron irradiated samples were tested at room-temperature (RT), in air, and at 1073 K in a helium atmosphere (total pressure 1 atm) with residual amounts of oxygen and hydrogen (100 appm) whilst helium-implanted bars tests were done at RT.

3. Results

Preliminary observations of neutron-irradiated specimens evidenced modification in the morphology of the material. Along the samples edges, perpendicular to the plane of the fibrous woven cloth, a fibre/matrix mismatch was visible using Scanning Electron Microscopy (SEM). These surfaces were pointed with cylindrical holes located along the original positions of fibre ends observed for non-irradiated materials (Fig. 1(a)). No visible dependence on the damage dose (1.29, 2.69 or 5 dpa) was found. Such effect is also observed in helium implanted materials (Fig. 1(b)), though in this case the mismatch is qualitatively more pronounced.

The results for flexure testing are presented in Table 1. Elastic modulus was obtained considering the initial linear zone of load-displacement curves and stresses were calculated using elastic beam theory. Application of this model to composites have a limited validity, in particular, following onset of matrix cracking, but provides useful qualitative data to discuss the behaviour of the material.

An average strength of 305 MPa is observed for the SiC/SiC composites in the "as-received" condition. Post-irradiation testing at RT for the 2500 appm helium implanted samples lead to a mean 38% decrease in strength down to values between 147 and 206 MPa. Force-displacement curves for these former cases are displayed in Fig. 2. An additional curve for bending bars annealed in helium at 1173 K is also displayed, in order to evaluate the effects of the temperature and atmosphere typical in the implantation experiments. From the curve for the implanted material, a decrease on the displacement at maximum load is evident. After this point, a damage tolerant behaviour is present, still composite toughness is lower, when compared with those observed to the non-implanted bars. In Fig. 3, a SEM image of the fracture region of one sample show an increased slip length of the fibre in the matrix, following matrix/fibre detachment.

Fig. 4 shows nominal stress-displacement curves for neutron irradiated materials, at room temperature and at 1073 K. Progressive strength degradation is observed as damage dose increases, when compared with RT results for the "as-received" material. Regardless of neutron dose, mechanical properties for the high-temperature tests are superior to those from RT-tests, as can be confirmed from the mechanical data in Table 1. At lower fluences (1.34×10^{21} n/cm^2), there is an improvement in toughness. As for 2500 appm He implanted materials, there is a pullout tail after σ_{Max}, but with the main difference that larger values for the displacement are attained. These large displacements are no longer present at higher fluences. The composite exhibits a brittle behaviour, with a slight recovery of the elastic modulus.

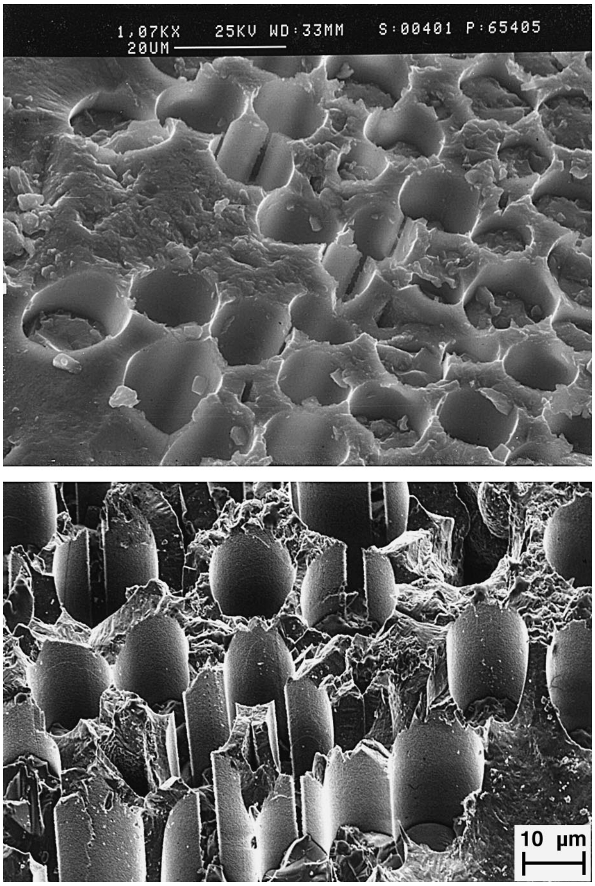

Fig. 1. Lateral surfaces of SiCf/SiC composites: (a) Neutron irradiated (accumulated damage dose – 1.29 dpa); (b) 2500 appm Helium Implanted (accumulated damage dose – 0.4 dpa)

4. Discussion

Stress-deflection curves for ceramic matrix composites are characterised by an initial linear zone followed by a non-linear regime until ultimate stress. This behaviour is greatly dependent on the characteristics of the fibre/matrix interface. Convenient tailoring of this phase in the composite permits multiple matrix cracking, debonding of the fibres from the matrix, fibre sliding and pullout, to ensure improved toughness of the material. It has been shown that a carbon interface thickness of 0.1 µm, as the one considered in our study, optimise composite strength whilst an acceptable toughness is also present [11].

For low fluences neutron irradiated materials, which in our case corresponds to the 1.34×10^{25} n/m^2, a density increase for the Si–C–O fibres has already occurred as reported in [12]. In the composite, the fibres are detaching from the matrix, and extensive fibre pullout leads to a progressive load-drop after maximum strength [13]. For the medium neutron fluences, corresponding to an accumulated damage of 2.69 and 5.23 dpa, contribution of these effects are no longer valid, as brittle behaviour arises, evidenced in the toughness reduction observed for the stress-displacement curve. Modifications of the fibre, matrix and interfaces properties have been reported, revealing densification and increase of elastic modulus for the fibres [12], and a decrease in the elastic modulus of the matrix [14]. Moreover, interfacial shear stress following an initial decrease from RT to low fluence (around 1 dpa) neutron irradiated materials, increases slightly as fluence increase [13,15]. These former parameters coupled with degradation of the fibre accounts for the composite embrittlement at medium and high neutron fluences. Whilst the SiC matrix response to neutron irradiation can be established from the results on monolithic pyrolitic beta-SiC [16], revealing a moderate swelling (linear expansion $<0.27\%$ for $T > 873$ K) mainly due to creation of point defects, microstructure changes induced by irradiation on the fibre phases are not clearly established.

Table 1
Three-point bending tests results on SiC_f/SiC (CVI) materials

Condition	Test temperature	σ_{MC} (MPa)	σ_{Max} (MPa)	d_{Max} (mm)	E (GPa)
"*As-received*"	RT	115	297	0.91	234
		122	339	0.95	238
		113	315	1.16	213
		129	304	0.83	227
		125	278	0.8	183
		128	304	0.81	214
36 h He	RT	119	275	0.94	239
1173 K		112	281	0.99	261
2500 appm He	RT	62	170	0.67	208
1173 K		93	178	0.73	153
		74	145	0.54	189
		90	206	0.72	217
Neutron 1.29 dpa	1073 K	111	242	1.79	128
1023 K		126	223	1.51	131
		108	240	1.7	120
	RT	63	196	1.56	110
		46	151	1.19	115
Neutron 2.69 dpa	1073 K	66	202	1.17	151
1023 K		117	223	1.29	136
		69	201	1.27	170
	RT	66	162	0.64	150
		131	160	0.6	130
Neutron 5.23 dpa	1073 K	58	154	0.9	107
1023 K		81	200	0.78	154
		100	180	0.8	131
	RT	95	160	0.8	117

σ_{MC} – Stress at first matrix cracking; σ_{Max} – maximum bending strength; d_{Max} – deflection at maximum strength; E – elastic modulus

For α-implanted material, the accumulated damage of ~0.4 dpa indicates that densification of the fibre already must have occurred [17]. Additionally, as for low fluence neutron irradiated materials, the fibres are likely to retain most of its strength after α-implantation. This latter statement was further strengthened by observing the maximum strength of unidirectional SiC/SiC composites, produced by a liquid impregnation/polymer pyrolysis route, remained almost unchanged after similar implantation doses [18]. Though stress displacement curves show similar features as those observed for 1 dpa neutron irradiated composites, a significant decrease of the values of d_{Max} is observed. Such behaviour indicates that toughening mechanisms after first matrix cracking (multiple matrix cracking; debonding of fibres; and friction between fibres and matrix) become less efficient following α-implantation. These toughening mechanisms are dependent on parameters such as interface bonding strength, residual radial stresses at the interface and fibre/matrix shear stress. These parameters should have values low enough to ensure that matrix cracks deflect along the interfaces, but sufficiently high to ensure that a relevant amount of fracture energy is dissipated. A possible explanation for the observed degradation is the swelling induced in the CVI β-SiC matrix by the implanted helium dose as discussed in [9]. This significant swelling leads to a wider fibre/matrix gap, helium-induced void formation at the interfaces, and even a pre-stress on the fibre-architecture due to the elongation of the fibre-"tunnels" in the matrix. Mainly the matrix being degraded by the implanted helium would also explain the lower onset of the deviation from linearity in the stress-displacement curves (so-called first matrix cracking). As the Nicalon CG-fibres are not significantly degraded, the value for σ_{Max}, which is only dependent on the fibre bundle – strength and – pullout, appears similar to those found for low fluence neutron irradiated materials. The results on helium implanted materials were obtained with the implantation zone geometrically limited within the whole sample volume. However, pre-stress effects on the fibres caused by a differential swelling of the matrix and fibre shrinkage

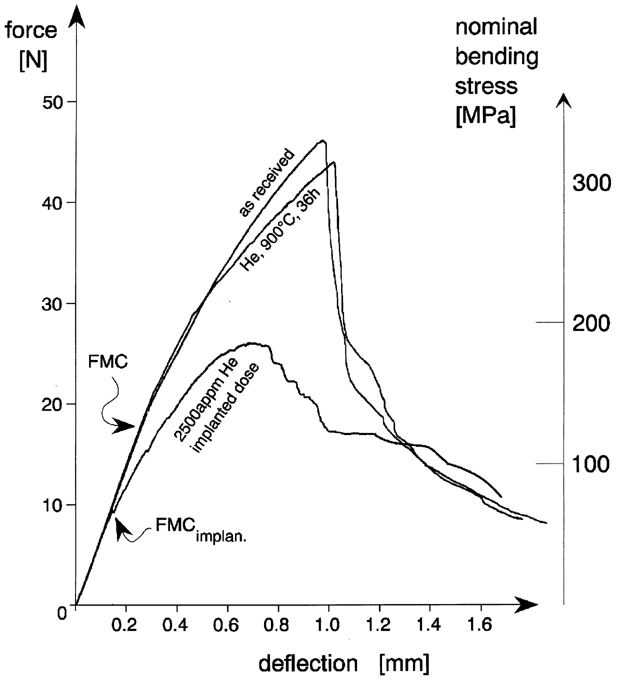

Fig. 2. Force-deflection curves for SiC/SiC composites – samples conditions are "as-received"; annealed in helium gas for 36 h at 1173 K and 2500 appm helium implanted.

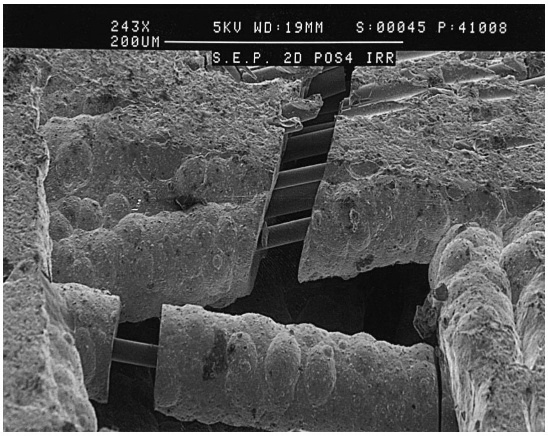

Fig. 3. SEM image of a fracture region in helium implanted SiC/SiC composites.

will influence the mechanical performance even in larger components homogeneously submitted to fusion rector relevant radiation.

5. Conclusions

For neutron irradiated materials, degradation in strength is occurring with increasing fluence and for damage doses higher than 2.69 dpa the composites exhibits a brittle behaviour. Degradation of the fibre due to irradiation (densification, microstructure evolution) accounts for the features in the composite mechanical behaviour. Mechanical tests at 1073 K show an improvement on the mechanical properties of the irradiated materials due to thermal recombination of defects. Still, Nicalon CG reinforcement is unlikely to be an acceptable reinforcement for a radiation resistant composite.

On the other hand, degradation of mechanical properties of helium implanted materials are attributed to swelling induced by helium in the CVI matrix coupled to Si–C–O fibre densification, i.e. differential swelling. Though microstructure processes accounting for this differential swelling effects are not yet fully established, their consequences on the mechanical behaviour of the composites becomes clear. Radial detachment of the swelled SiC (CVI) matrix modifies the fibre/matrix interface in the composite, thus limiting the synergetic load share/transfer between the ceramic matrix and the fibrous reinforcement. As a consequence, decrease in strength and displacement at maximum load is observed for the implanted materials under flexure loading. Data for helium effects on SiC, covering different helium doses in a temperature range from RT up to 1273 K, is limited. Future studies should improve this data, so that a deeper understanding of the behaviour of polycrystalline SiC,

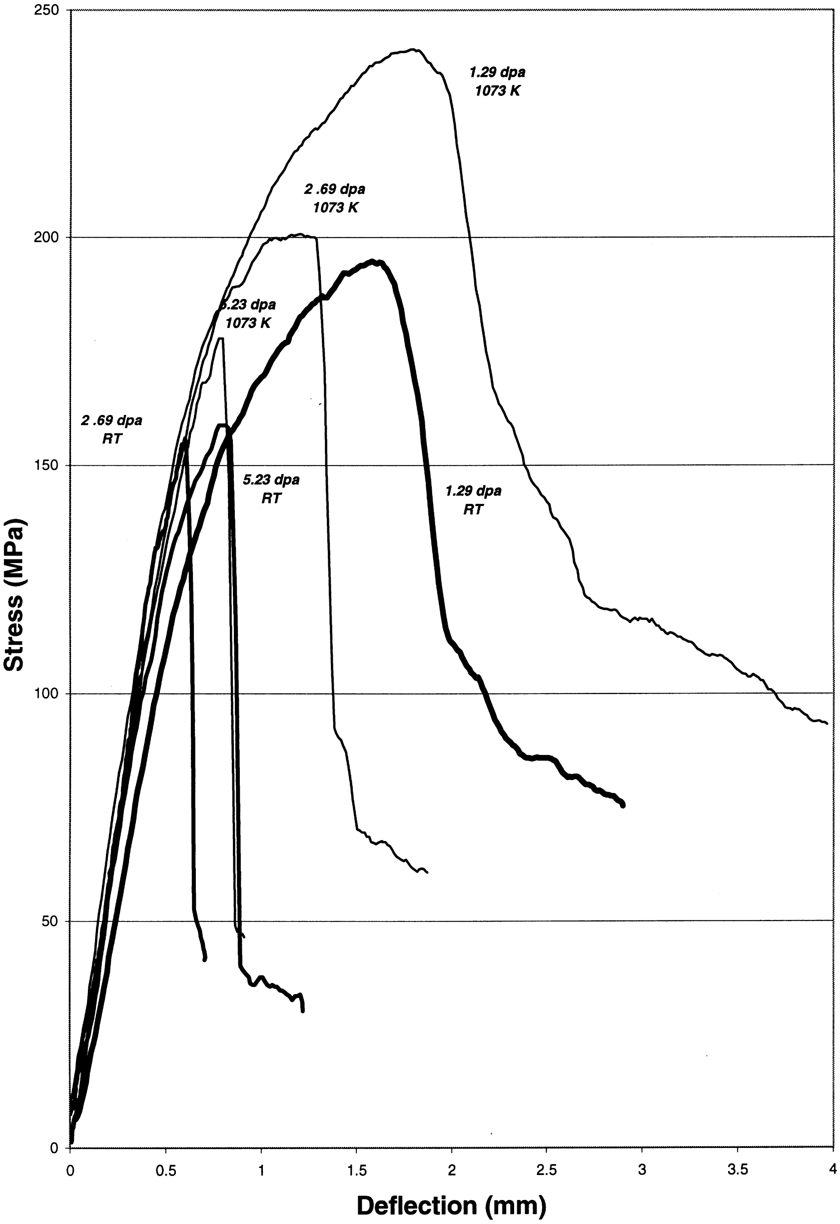

Fig. 4. Stress-displacement curves, at RT and 1073 K, for neutron irradiated SiC_f/SiC composites.

containing fusion relevant helium quantities, could be established.

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