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Silicon carbide fibre-reinforced glass-ceramic composite tensile behaviour at elevated temperature

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The tensile performance of 0/90 cross-ply composite of SiC fibre-reinforced glass-ceramic is examined as a function of temperature, environment and stressing conditions. Tension, fatigue and stress rupture tests were all performed in both air and argon atmospheres up to a maximum temperature of 1300° C. It is shown that, in argon, composite ultimate strength is retained up to 1300° C. In air, strength loss occurs at elevated temperatures with an associated change in failure mode. Prolonged loading in fatigue or under constant stress in air at high temperature causes further strength loss which appears limited by the proportional limit stress of the composite.

1. Introduction

Silicon carbide-type fibre-reinforced glass-ceramic matrix composites have been developed to meet the need for low-density fracture tough materials to replace metals in high-temperature applications. The creation of a Nicalon fibre-reinforced lithium-aluminium-sulphate (LAS) matrix composite clearly demonstrated the potential for this type of composite, both in terms of high strength and fracture toughness [1]. In this initial development, flexural tests were performed in an inert argon atmosphere at temperatures of up to 1200° C with both strength and toughness being maintained at up to 1100° C Further investigation of this composite resulted in the discovery that its strength and toughness are, in large measure, due to the existence of a continuous carbon-rich layer at the fibre–matrix interface [2]. This interface is apparently created at the high temperatures of composite fabrication through the instability of the Nicalon fibre while the fibres are in intimate contact with the LAS matrix. The creation of this interface is also accompanied by a significant reduction in fibre tensile strength, which also relates to the particular matrix composition being used. Extracted fibre strengths were related to composite tensile strength and stress–strain behaviour and it was found that composite failure strains could significantly exceed those of matrix cracking when the extracted fibre strengths were high [3, 4]. The existence of this cracking, however, permitted the surrounding atmosphere to enter into the composite structure. When tests were performed at elevated temperature in air [5, 6], it was found that these microcracks permitted the ingress of oxygen into the fibre–matrix interfacial region, degrading the interface through the removal of the carbon-rich zone. This effect was found to be particularly pronounced when applied stresses exceeded the composite's proportional limit stress (strain), i.e. the stress (strain) at which microcracks form extensively in the matrix.

It was the purpose of the present work to examine composite tensile stress–strain behaviour in a series of tests in which loading was in pure tension to minimize any stress state complexities and determine the effects of test temperature and test environment on tensile strength. To minimize testing difficulties, all specimens had fibres oriented in both the 0° and 90° directions, This decreased the tendency for axial shear along the fibres and also reduced the load that the specimens would fail at, to minimize gripping problems. In addition, the cross-ply fibre orientation is more closely related to composites that might actually be used.

2. Experimental procedure

2.1. Tensile specimen configuration and test procedure

The tensile specimens were each machined to the configuration shown in Fig. 1. This specimen configuration had three 0.3 cm diameter holes that were used for holding the head of the specimen during loading. The specimen gauge area was 2.5 cm long and 1 cm wide. A 5 cm radius was used for the smooth transition from the gauge section to the head width.

This tensile configuration worked well for temperatures up to 1100° C. At 1300° C the retaining pins pulled through the head section of one specimen at a gauge section stress of 430 MPa. This specimen was the first tested over 1100° C The one remaining specimen at 1300° C and the two specimens at 1200° C were reduced to a gauge section width of 0.7 cm. The gauge

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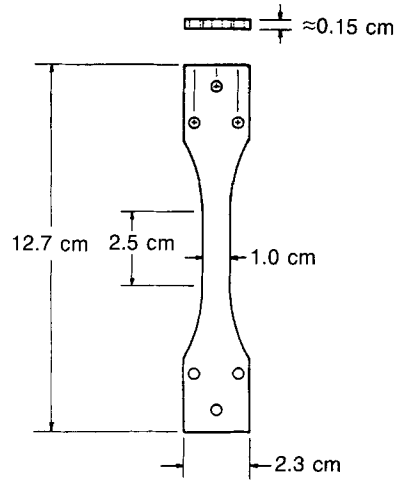

Figure 1 (0/90) SiC/LAS tensile specimen configuration.

section width reduction decreased the load on the specimen pin holes and allowed proper tensile failure in the gauge section.

The high-temperature tensile evaluations were performed in a gas-bearing tensile facility. This apparatus utilized gas-bearing universals in the load linkage to prevent the introduction of unknown bending moments in the load train from cross-head motion or eccentricity and to allow monitoring of the load train. For tensile testing, loads were applied at 70 MPa min^{-1}.

Strain measurements were made with clip-on extensometers. Ports on opposite sides of the furnace provided a means of attaching two sets of clip-on arms to the specimen at a predetermined gauge length of 2.2 cm. For the air atmosphere tests the clip-on arms were attached after the specimen had reached the test temperature and then allowed to stabilize for a few minutes.

Specimen temperatures to 1000° C were determined by thermocouples placed at the centre of the gauge section; optical pyrometers wre used to measure temperature above 1000° C.

2.2. Composite material

A high-temperature LAS glass-ceramic matrix was used. Designated LAS-III, this composition is similar to a Corning Glass 9608 LAS except that it has been tailored to be more compatible with SiC-type fibres. The glass was obtained in the form of fine powder which was then used to make composites according to the hot-pressing techniques described previously.

The fibre utilized was Nicalon (Nippon Carbon Co., Yokohama, Japan) having an average elastic modulus of 190 GPa, tensile strength of approximately 2000 MPa and a density of 2.5 g cm^{-3}. The fibre was utilized to make unidirectionally aligned tapes. Composites were fabricated by the hot-press densification of plys of these tapes. In the case of these ply lay-up composites, it will be noted that two different types of fibre orientation were used. The specimens designated $[0(0/90_2]_s$ were 10 plys thick and were made using unidirectional fibre plys laid up in the indicated symmetrical fashion. These composites contained extra 0° plys on the outer surfaces due to initial experiences where it had been necessary to grind the outer composite surfaces prior to testing. In this present study, however, no surface grinding was performed and the surface plys were retained intact. Other composites designated simply by the $[0/90]_{4s}$ ply sequence were 8 plys thick and did not have extra 0° plys on their surface. All composites contained approximately 40 to 45% by volume fibre.

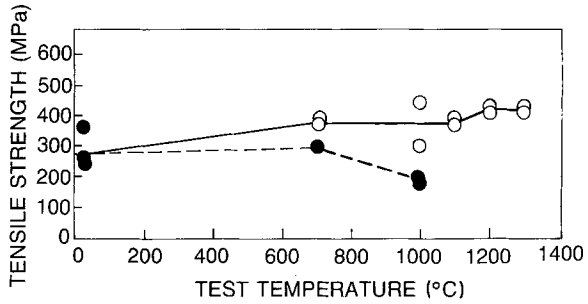

Figure 2 Tensile strength of $[0(0/90)_2]_s$ SiC/LAS-III tested in (●) air and (○) argon.

3. Experimental results

3.1. $[0(0/90)_2]_s$ SiC/LAS-III composite tensile tests

The average tensile properties at each temperature are summarized in Table I along with an indication of the appearance of the resultant fracture surfaces. The tensile strengths are plotted as a function of temperature in Fig. 2 where it can be seen that at 700 and 1000° C the strength in air is substantially reduced. Examination of the composite fracture surfaces, however, revealed only small differences in fracture morphology due to the test atmosphere. As seen in Fig. 3, specimens tested at 1000° C were fibrous in failure mode for both atmospheres. The only noticeable difference was that a thin embrittled region could be noted right at the specimen surfaces. The only slight change in failure mode may be associated with the fact that the specimens were loaded at a rate of 70 MPa min^{-1}. The longer term stress rupture

TABLE I Tensile properties of $[0(0/90)_2]_s$ SiC/LAS-III composites

	Temperature (° C)							
	20	700		1000		1100	1200	1300
Atmosphere	Air	Air	Argon	Air	Argon	Argon	Argon	Argon
Strength (MPa)	269	307	382	201	375	380	425	424
Modulus (GPa)	75.8	100	71.1	99.2	88.9	75.8	80.6	68.9
Strain-to-failure (%)	0.5	0.67	0.88	0.62	1.11	0.9	1.15	1.07
Fracture mode*	F	F	F	SF	F	F	F	F

*F = fibrous fracture, SF = only short fibre lengths visible. Poisson ratio values of 0.085, 0.075, and 0.08 were measured from the room-temperature specimens.

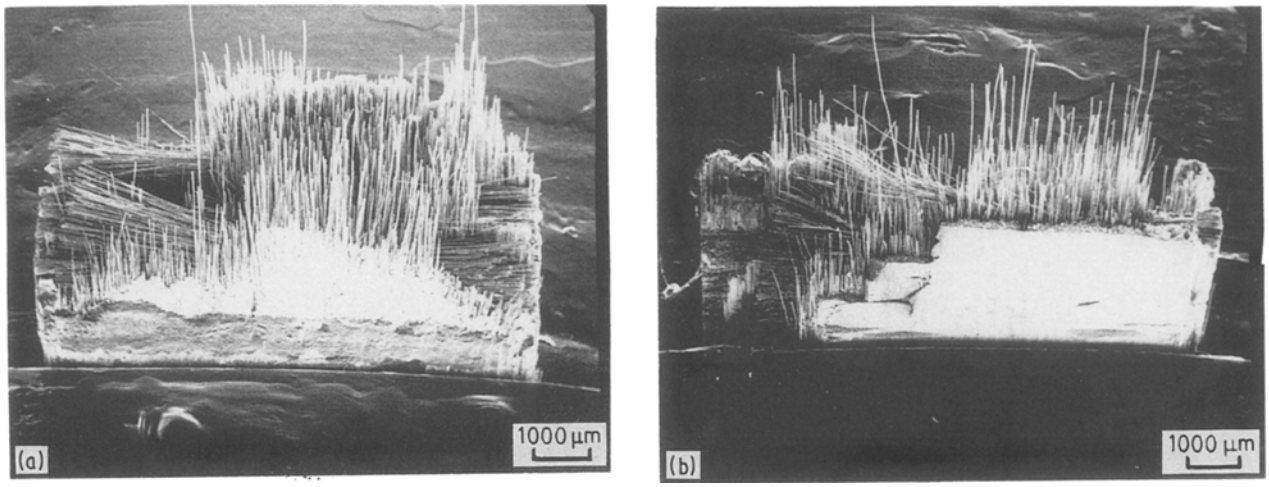

Figure 3 Fracture surfaces of $[0(0/90)_2]_s$ SiC/LAS-III tensile specimens tested at 1000° C. in (a) argon and (b) air. UTS: (a) 300 MPa, (b) 225 MPa.

and fatigue tests to be described later exhibited more marked effects.

The high strength and fibrous fracture mode obtained at 1300° C (Fig. 4) are particularly notable in demonstrating the high-temperature potential of these composites. By ascribing zero strength to the 90° oriented plys it was possible to calculate the effective 0° ply strength in argon for this composite (Fig. 5). It can be seen that strength increases with test temperature and approaches 700 MPa at 1300° C. This is a rather remarkable finding in that is was aniticipated that 1200° C and above the fibres would rather rapidly lose their strength. It would appear that once encapsulated in the LAS matrix, at least for short times, this is not the case.

Composite stress–strain curves are presented in Figs 6 to 8 where it can be seen that behaviour is quite nonlinear. At room temperature (Fig. 6), several different regions of stress–strain response can be noted. At 38.6 MPa the first transition occurs due to matrix cracking or fibre–matrix interfacial debonding in the 90° plys and at approximately 175 MPa pronounced evidence of similar effects in the 0° plys is noted. In the past [6], it has been useful to refer also to a 0.02% proportional limit stress and in this case this stress occurs at 110 MPa, well below the pronounced "knee" in the curve. The 0.02% limit, which is taken at a strain off-set of 0.2%, is useful in that it expresses a stress associated with a standard of deviation from linear behaviour that may eventually be useful for design purposes. At higher temperatures (Figs 7, 8), nonlinearity is preserved in all of the curves and at 1300° C composite failure strain exceeds 1%.

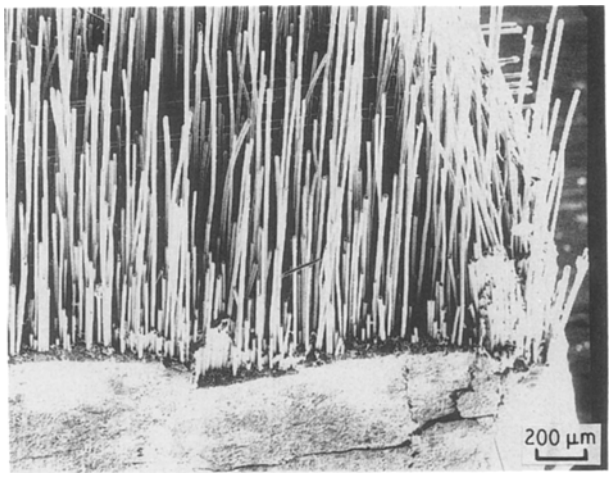

Figure 4 Fracture surface of $[0(0/90)_2]_s$ SiC/LAS-III tensile tested in argon at 1300° C. Ultimate tensile strength = 420 MPa.

3.2. $[0/90]_{4s}$ SiC/LAS-III composite tension–tension fatigue at 900° C

Tension–tension fatigue tests were performed with a ratio of minimum to maximum applied stress of 0.1. All of these specimens were fabricated using a balanced (0/90) ply lay up that did not include extra 0° plys on the surface. For this reason composite tensile strengths were less than those described in the above section. Tensile tests were performed first at 900 and 1100° C and then cyclic fatigue conditions were selected for further testing at these temperatures. The tensile stress–strain curves, obtained in air are shown in Fig. 9.

The rate of fatigue cycling varied from between 7.2 and 10.0 c.p.s. and composite elastic modulus was measured by performing a slower loading cycle at a rate of 70 MPa min^{-1} on the 1st, 1000th, 10 000th and 100 000th cycles.

The results of fatigue testing at 900° C in air are summarized in Fig. 10. While the composite tensile strength at this temperature was 150 MPa, composite fatigue performance was limited to an applied stress of approximately 86 MPa. This stess level was equivalent to the residual strength of two specimens prefatigued

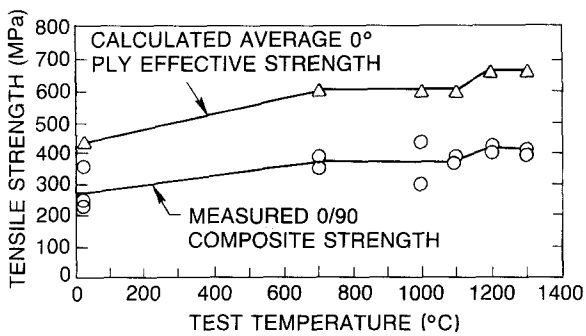

Figure 5 Tensile strength in argon of $[0(0/90)_2]_s$ SiC/LAS-III.

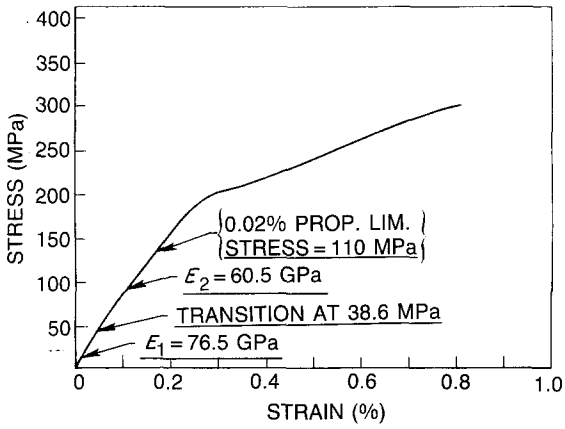

Figure 6 Tensile stress–strain curve for $[0(0/90)_2]_s$ SiC/LAS-III at 22° C.

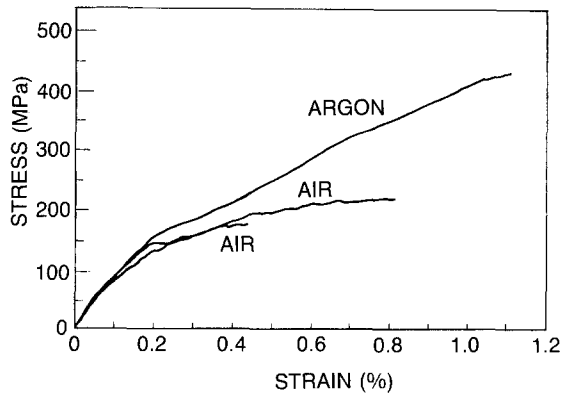

Figure 7 Tensile stress–strain curves for three specimens of $[0(0/90)_2]_s$ SiC/LAS-III tested at 1000° C.

to 100 000 cycles under an applied stress of 69 MPa and also the stress level which caused the failure of specimens at 340 000 and 440 000 cycles. Referring to Fig. 9 it is possible to see that this limiting stress condition is in close agreement with the stress at which these composites passed through the proportional limit and probably evinced extensive microcracking at 900° C.

As mentioned above, these fatigue tests were periodically interrupted to measure composite elastic modulus to ascertain if composite damage was occurring with increasing numbers of test cycles. The data, presented in Table II, indicate that in all cases composite stiffness was unaffected by fatigue. If significant amounts of matrix cracking or fibre debonding had been occurring it would have been evinced by a loss in stiffness. This, however, was clearly not the case.

A comparison of tensile and fatigue-induced failure surfaces in air was made (Fig. 11). Both fractures appear fibrous although the length of the fibre pull-out is slightly less for the fatigue specimen which required 16.9 h to fail during testing.

In marked contrast, fatigue tests performed in argon were not limited by the proportional limit stress (Fig. 12). The residual tensile strength of specimens fatigued to 100 000 cycles was equivalent to the strength of specimens which had been simply tensile tested without any prefatigue (Fig. 12). Two additional specimens failed during fatigue testing when stressed at levels of 140 and 100 MPa. These stress levels are very close to those chosen for the specimens which exhibited the high residual stresses described above. This observation implies the existence of a rather small difference between conditions which can cause composite fatigue failure and those which do not. This is analogous to the results obtained by testing similar composite samples in tensile fatigue at room temperature [7]. In this work it was found that specimens which survived 100 000 cycles of fatigue testing at 22° C exhibited retained tensile strengths equivalent to unfatigued composites. A major difference between 22° C and high-temperature findings, however, was that the applied fatigue stress levels at 22° C were significantly higher than those at 900° C.

3.3. $[0/90]_{4s}$ SiC/LAS-III composite tensile stress rupture testing at 900 and 1100° C

Tensile stress rupture testing in air at 900° C indicated a 100 h survival for applied stresses of between 48 and 70 MPa (Fig. 13). In contrast, testing in argon (Fig. 14) indicated a 100 h rupture strength of 105 MPa or higher, indicating once again the strong effect of environment. At 1100° C in air, specimens stressed at 48 MPa survived up to 162.3 h and exhibited residual strengths of approximately 117 MPa (Fig. 15). This is greater than the residual strength of similar specimens tested in air under the same 48 MPa but at 900° C, indicating a potentially lesser effect of environment at the higher temperature, presumably due to the ability of the glass-ceramic to deform more extensively. A comparison of fracture surfaces confirms this hypothesis in that the specimens tested at 900° C exhibited almost no fibre pull-out while the 1100° C specimen is embrittled only at its outer surfaces.

The much more significant effect of environment on composite fracture appearance for stress rupture tests (Fig. 16), as compared to fatigue tests (Fig. 11), probably relates to the much longer time under stress in the stress rupture tests. The fatigue tests were completed in 16.9 h while the stress rupture tests in air ran for up to 144 h. The degradation of these composites is clearly time dependent.

TABLE II Composite elastic modulus as a function of number of fatigue cycles at 900° C in Air; $[0/90]_{4s}$ SiC/LAS-III

Applied fatigue stress (MPA)	Elastic modulus (GPa) at indicated cycle number					Total number of cycles	Retained tensile strength (MPa)
	1	2	1000	10 000	100 000		
69	69	64	62	64	64	100 000	85
69	82	90	80	80	76	100 000	86
86	73	67	65	64	66	339 860	–
86	73	73	70	72	72	438 830	–

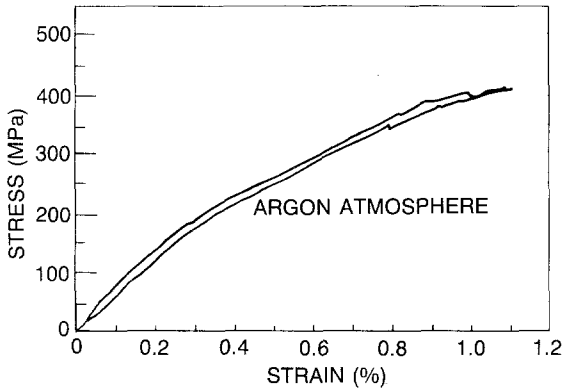

Figure 8 Tensile stress–strain curves for two specimens of $[0(0/90)_2]_s$ SiC/LAS-III tested at 1300° C in argon.

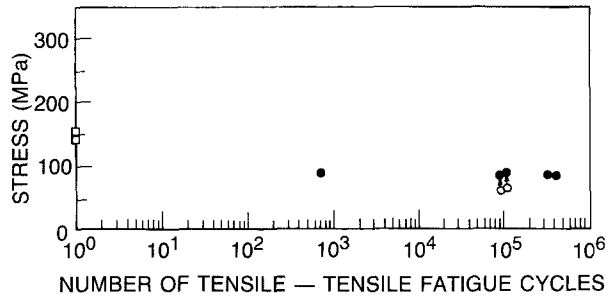

Figure 10 Tension–tension fatigue data for $[0/90]_{4s}$ SiC/LAS-III specimens tested at 900° C in air. (□) Monotonic tensile data, (○) maximum tensile fatigue stress without failure, (●) maximum tensile fatigue stress with failure. Arrowed (●) retained tensile strength.

4. Conclusion

The tensile behaviour of these multiaxially reinforced composites is quite complex. As shown in Fig. 6, the stress–strain curve is highly nonlinear with several discernable transitions in shape. The initial application of stress resulted in an elastic modulus of 76.5 GPa. However, at a stress of 38.6 MPa it was possible to detect a reduction in stiffness resulting in a modulus of 60.5 GPa. This decrease can be most readily associated with the loss of 90° ply contribution to the overall composite elastic modulus. It was previously shown that the 90° ply elastic modulus is approximately 40 MPa and the failure strain is 0.04% for composites of this type [3]. For the ply lay-up associated with Fig. 6, 40% of the plys are in the 90° orientation and hence a loss of their contribution would account for 16 GPa reduction in stiffness which is in very close agreement with the value observed. The next major transition in the curve occurred at a stress of approximately 175 MPa (strain of 0.25%). This transition appears to be very similar to that observed for uniaxially reinforced composites [3, 4] and has been associated with the occurrence of a large amount of microcracking in the matrix of the 0° oriented plys.

This overall tensile stress–strain curve shape was generally retained to high temperatures when tested in argon. Only at 1300° C (Fig. 8) was it altered significantly such that no abrupt transition in slope could be detected. At this highest temperature the matrix failure strain was probably much higher than at the lower temperatures and hence matrix failure did not occur as precipitously. Also noteworthy is the retention of modulus and strength of these composites at up to 1300° C. This was somewhat unexpected in that the fibres are initially produced at a maximum temperature of less than 1300° C, and high-temperature testing of fibres alone has shown property loss at these temperatures [8]. Composite processing at high temperatures causes a significant loss in apparent fibre strength, as determined by leaching fibres out of the composites [3], however, this subsequent tensile test exposure to high temperature has not been detrimental. The reason for the high strength at 1300° C may also be due to a change in failure process; however, the fractograph shown in Fig. 4 appears to be very similar to that characteristic of specimens tested at 22° C.

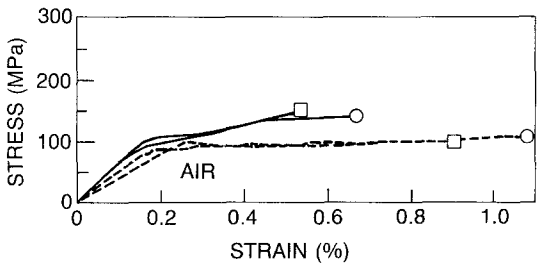

Figure 9 Tensile stress–strain curves for two specimens of $(0/90)_{4s}$ SiC/LAS-III tested at (——) 900° C and two specimens tested at (– – –) 1100° C in air.

The effect of test atmosphere was very pronounced. By tensile testing in air the composites were limited to a strength approximately equivalent to that associated with the 0° ply proportional limit stress. This is most apparent from composites tested at 1000° C (Fig. 7), and agress with the concept of extensive matrix cracking providing the opportunity for air to enter the fibre–matrix interfacial region to degrade composite performance [5, 6]. The tests which aggravate this situation through the application of cyclic stress or prolonged constant stress illustrate this point even more clearly. In both of these cases, the stress limit associated with the 0° ply cracking provided the lower limit for composite performance and those tests performed over the longest time period caused the greatest change in fracture morphology. Prolonged exposure under stress caused marked reduction in the amount of fibre pull-out even if the applied stress was less than that associated with matrix cracking. It was also apparent that the degradation process propagated inward from the specimen surfaces, even at low applied stress.

It can be concluded that the use of Nicalon fibres can provide these glass-ceramic matrix composites with exceptional strength at temperatures of up to 1300° C. The inability to retain this strength in an oxidizing environment, however, is dependent on the time of exposure and also the stress at which extensive microcracking in the composite occurs. Experiments which measured the fibre–matrix interfacial bond strength at both room temperature and 1000° C in air [9], have confirmed that the oxidizing environment apparently alters the nature of the fibre–matrix bond and causes matrix cracks to propagate into the fibres

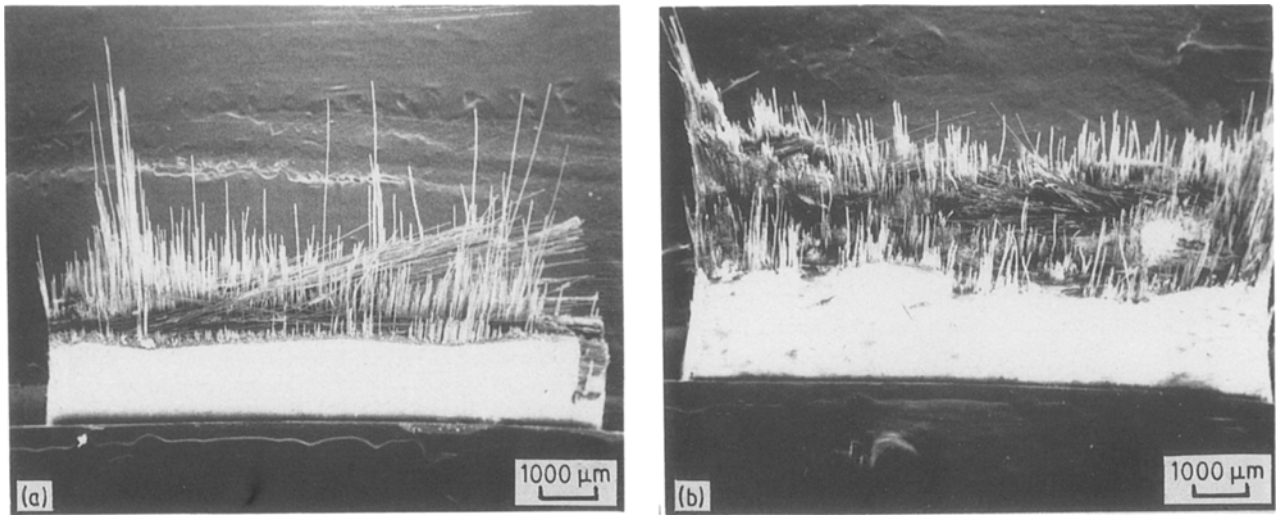

Figure 11 Fracture surfaces of $[0/90]_{4s}$ SiC/LAS-III specimens tested to failure in tension and tension–tension fatigue at 900° C in air. (a) Tensile test, UTS = 153 MPa; (b) failure after 438, 830 cycles at 86 MPa.

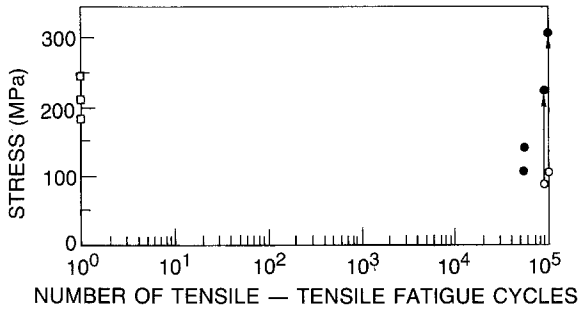

Figure 12 Tension–tension fatigue data for $[0/90]_{4s}$ SiC/LAS-III specimens tested at 900° C in argon. (□) Monotonic tensile data, (○) maximum tensile fatigue stress without failure, (●) maximum tensile fatigue stress with failure. Arrowed (●) retained tensile strength.

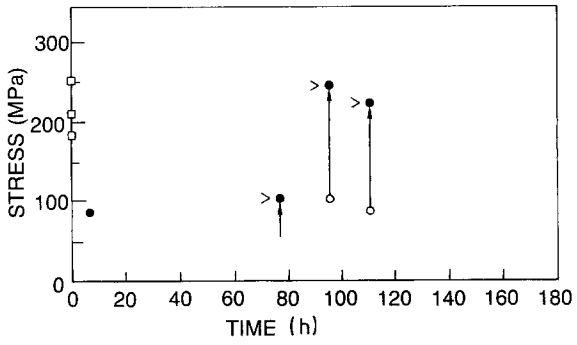

Figure 14 Tensile stress rupture data for $[0/90]_{4s}$ SiC/LAS-III specimens tested at 900° C in argon. (□) Monotonic tensile data, (○) constant tensile stress without failure, (●) constant tensile stress with failure. Arrowed (> ●), retained tensile strength greater than this value due to improper failure.

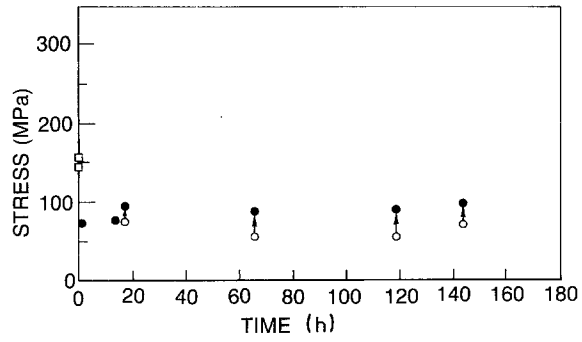

Figure 13 Tensile stress rupture data for $[0/90]_{4s}$ SiC/LAS-III specimens tested at 900° C in air. (□) Monotonic tensile data, (○) constant tensile stress without failure, (●) constant tensile stress with failure. Arrowed (●) retained tensile strength.

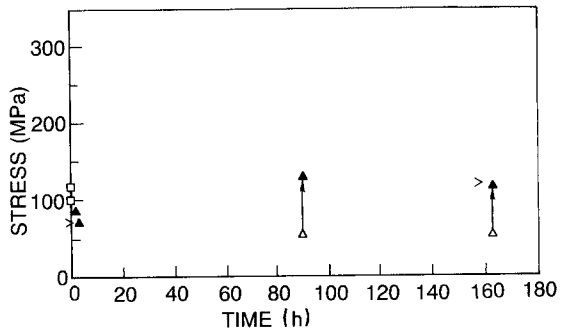

Figure 15 Tensile stress rupture data for $[0/90]_{4s}$ SiC/LAS-III specimens tested at 1100° C in air. (□) Monotonic tensile data, (△) constant tensile stress without failure, (▲) constant tensile stress with failure. Arrowed (▲), retained tensile strength, arrowed (> ▲) retained tensile strength greater than this value due to improper failure.

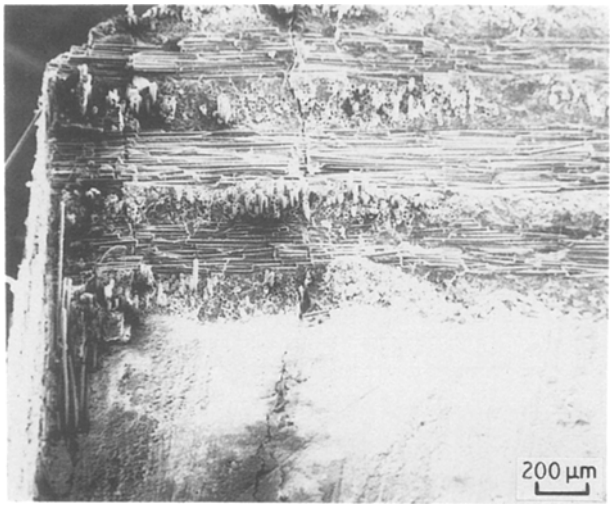

Figure 16 Fracture surface of $[0/90]_{4s}$ SiC/LAS-III tensile stress rupture specimen exhibiting a residual tensile strength of 95 MPa after 144 h exposure to an applied stress of 69 MPa in air.

causing a significant decrease in both composite strength and apparent toughness after prolonged exposure.

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