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Stress-Rupture of Nicalon/SiC Continuous Fiber Ceramic Composites in Air at 950°C

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The stress-rupture behavior of plain-weave CG-Nicalon/enhanced SiC was studied in air at 950°C. It was found that this material exhibits delayed failure and that the lives of specimens subjected to constant stress levels of 80, 100, and 120 MPa were 21.6, 9.6, and 2.7 h, respectively. The strain histories of these tests revealed a continuous increase of the specimen compliance and accelerated deformation prior to failure. It is shown that both the shape of the strain vs time curves and the time dependence of the loss of strength can be explained using a simple model based on the oxidation-induced stress-rupture of the reinforcing fiber bundles.

I. Introduction

The driving force behind the development of continuous-fiber-reinforced ceramic composites (CFCCs) is the promise of substantial economic and environmental benefits if they are used in energy and related industrial technologies, particularly at elevated temperatures.[1] The main attraction for using CFCCs over other high-temperature materials, particularly monolithic ceramics, is their potential to retain strength, their superior toughness, and noncatastrophic mode of failure.

Since many of the potential applications for CFCCs involve components with lives that are measured in tens of thousands of hours,[1] the successful design and implementation of CFCC components will depend on the availability of life-prediction methodologies and knowledge of the material behavior over periods of time comparable to the expected service life of the component. These propositions reflect the motivation behind the work presented in this paper.

The ultimate strength and reliability of CFCCs are determined primarily by the reinforcing fibers and, to a lesser extent, by the fiber–matrix interface. The fiber–matrix interface plays a major role by providing stress transfer between the fibers and matrix, and by controlling the ability of the material to promote fiber debonding and fiber sliding in the wake of an advancing crack in the matrix. Typically, the engineering of the fiber–matrix interface in CFCCs relies on the use of fiber coatings.

The availability of SiC fibers and techniques for the densification of SiC matrices have made SiC/SiC composites a prime material candidate for many energy-related industrial applications. In many of these applications, subjecting CFCCs to stresses larger than the matrix cracking stress will result in the ingress of the environment, often oxidizing, to the interior of the composite. In the absence of oxidation-resistant fiber coatings this would result in the degradation of both the fiber coating and the fibers, leading ultimately to fiber and composite failure.[2–7] Therefore, it has become clear that the durability of CFCCs will be contingent upon how well the fibers can be protected from degradation, either from environmental attack and/or from reacting with other composite constituents.

Studies of the stress-rupture behavior of plain-weave CG-Nicalon/enhanced SiC have revealed that this material exhibits delayed failure when subjected to stresses larger than 55 MPa.[8] However, unlike what was stated in that paper, the changes in specimen compliance recorded during those stress-rupture tests are not associated with material creep (i.e., flow), but rather it now appears that they arise from the progressive breakdown of the reinforcing fiber bundles, as is shown below and elsewhere.[5–7] Studies of the stress-rupture behavior of CG-Nicalon/SiC in air have demonstrated that this class of materials exhibits delayed failure even at temperatures as low as 425°C.[5–7]

In this paper, experimental results are presented from a study of the stress-rupture behavior of CG-Nicalon/enhanced SiC CFCCs in air at 950°C. The phenomenology associated with these tests, namely increasing compliance with time, and delayed failure, is explained in terms of the oxidation-induced breakdown of the reinforcing fiber bundles.

II. Experimental Procedure

The material evaluated consisted of CG-Nicalon [0/90] plain weave fabric, coated with a layer of pyrocarbon prior to matrix densification. The SiC matrix was densified by chemical vapor infiltration (CVI) and contained an enhancement of proprietary composition.[9] The total volume fraction of fibers was 40%. The tensile specimens had dimensions shown in Fig. 1, and had an outer seal coating of SiC that was applied by chemical vapor deposition (CVD) after the specimens had been machined. The width of the gauge section contained three wavelengths of the fabric weave pattern.

Tensile stress-rupture tests were conducted per ASTM C1337[10] in ambient air (24–30% relative humidity) using an electromechanical test machine equipped with hydraulically actuated grips. Specimen deformation was monitored over a 25-mm-long gauge length using a low-contact force capacitance extensometer, and the specimens were heated using a compact resistance-heated furnace. The alignment of the load train was verified prior to each test using a strain gauged metallic test specimen per ASTM 1295.[11] The grip wedges were water-cooled to preserve the adhesively bonded aluminum end-tabs that were used to minimize specimen damage during gripping. The specimens were heated in ambient air from ambient temperature up to 950°C at a constant rate of 20°C/min, followed by a 20 min soaking period. At that point the specimens were loaded at a constant rate of 5 MPa/s up to the test load, which was maintained constant thereafter for the duration of the test. The tests were completed when the specimens failed.

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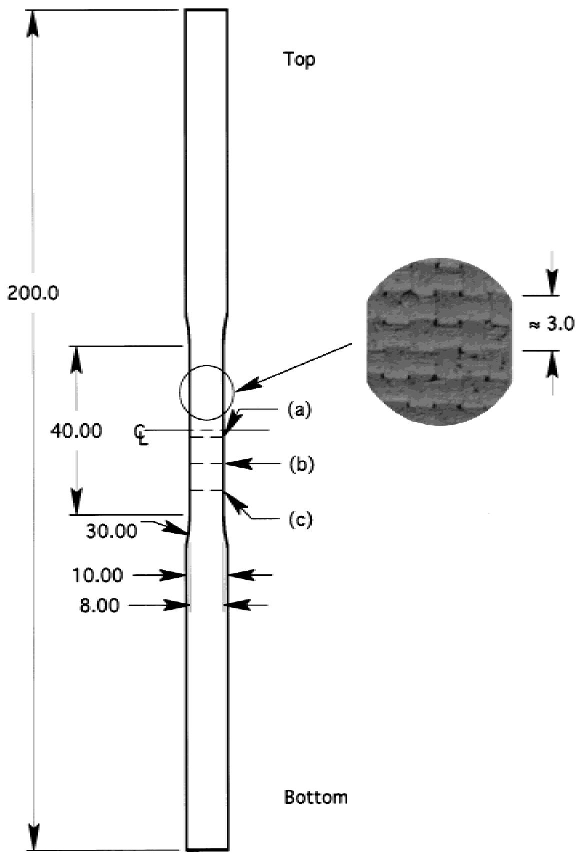

Fig. 1. Schematic of tensile specimen geometry and dimensions in mm. Also shown are the failure locations for the three tests conducted in ambient air at 950°C: (a) 80, (b) 100, (c) 120 MPa.

Failures occurred within the uniformly heated gauge section (≈35 mm long) of the specimen as indicated in Fig. 1.

III. Results

Figure 2 shows the strain histories of stress-rupture tests conducted in air at 950°C at stress levels of 80, 100, and 120 MPa. The resulting strain vs time curves revealed that the material became increasingly compliant with time, and that for stresses of 100 and 120 MPa, it exhibited accelerated deformation prior to failure. Figure 3 shows the stress vs strain curves associated with these tests and illustrates that in all three cases, the proportional stress limit (≈71.3 ± 1.1 MPa), which was larger than the matrix cracking stress, was exceeded. Also it was found that the slope of the initial linear region of the stress vs strain curves was equivalent to an elastic modulus of 116.7 ± 6.1 GPa.

Figure 4 shows a plot of the rupture stress as a function of the time-to-failure for the three tests. Also included in Fig. 4 are data from the evaluation of the same material in air at 982°C, although some of those specimens failed at a location outside of the uniformly heated gauge section where the temperature was lower than the test temperature.[8]

IV. Analysis and Discussion

In order to account for the shape of the strain versus time curves and the time dependence of the loss of strength obtained during the stress-rupture of CG-Nicalon/enhanced SiC in ambient air at 950°C, a very simple model based on the oxidation-induced stress-rupture of the reinforcing fiber bundles will be used.

Consider a bundle with a large but finite number (N_0) of fibers of uniform cross-sectional area, A, that have the same linear stress–strain curve. If it is assumed that the strength-controlling flaws are restricted to the surface of the fibers and that the strength of the fibers is described by a two-parameter Weibull distribution, then the probability of failure of the fibers when subjected to a uniform tensile stress T, will be given by

$$\Phi = 1 - \exp\left\{-\frac{\ell}{\ell_0}\left(\frac{T}{\overline{\sigma}_0}\right)^m\right\} = 1 - \frac{N}{N_0} \tag{1}$$

where ℓ is the fiber gauge length, $\overline{\sigma}_0$ is the Weibull characteristic strength, ℓ_0 is the reference gauge length, m is the Weibull modulus, and N is the number of surviving fibers. If the bundle behaves in the Daniels sense, i.e., that the load applied to the bundle is equally distributed among the surviving fibers (global load sharing),[12,13] the relationship between the applied load, F, and the stress on each fiber, T, will be given by

$$T = \frac{F}{N_0A(1-\Phi)} = \frac{F}{AN} \tag{2}$$

whereas the elongation of the bundle under a loaf F will be simply given by

$$\varepsilon = \frac{F}{NAE} = \frac{T}{E} \tag{3}$$

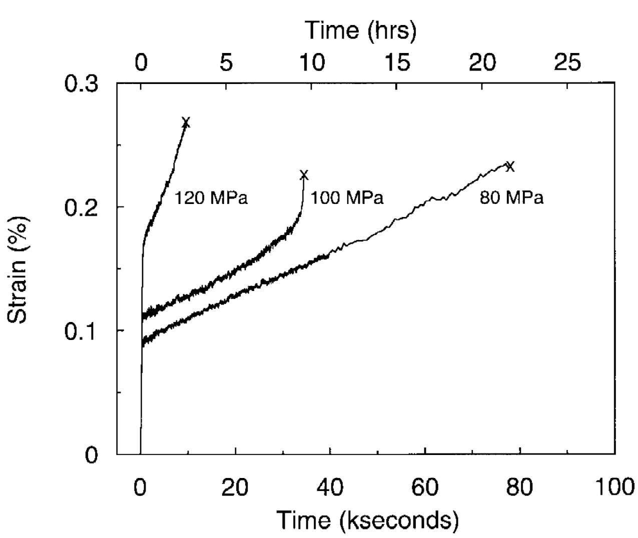

Fig. 2. Deformation histories recorded during the stress-rupture evaluation of Nicalon/enhanced SiC in air at 950°C.

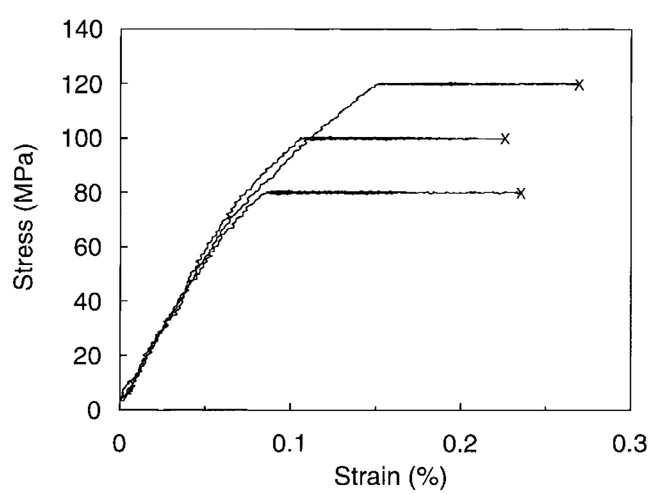

Fig. 3. Stress versus strain curves recorded during the stress-rupture evaluation of Nicalon/enhanced-SiC in air at 950°C.

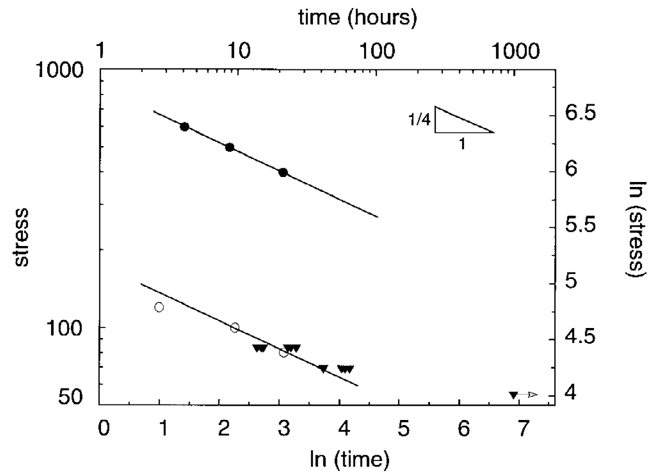

Fig. 4. Summary of stress rupture results: (○) this work, (▼) Verilli *et al.*,[8] (●) predicted stress rupture of Nicalon fiber bundle.

Let us consider a bundle of CG-Nicalon fibers that is subjected to a constant tensile load in ambient air at a temperature at which the fibers do not flow (i.e., do not creep), but do oxidize.[†] For simplicity it is assumed that both the Weibull and Young moduli of the fibers remain constant and that the only effect of oxidation is to decrease the characteristic strength of the fibers. In this case, the time-dependent strength of the fibers will be controlled by surface defects resulting from oxidation, with the thickness of the oxide layer representing the size of the average strength-controlling flaw.[14] Considering that the oxidation of CG-Nicalon and other SiC-based fibers is controlled by diffusion of oxygen through the oxide layer, the oxide layer will grow on the fiber's surface according to[‡]

$$\alpha = \sqrt{kt} \tag{4}$$

where α is the thickness of the oxide layer at time t, and k is the parabolic rate constant.[15,16]

According to linear elastic fracture mechanics, strength and critical flaw size are related through the fracture toughness as follows:

$$K_{\mathrm{IC}} = Y\sigma_0\sqrt{a} \tag{5}$$

where K_{IC} is the fracture toughness, Y is a geometric parameter, σ_0 is the strength, and a is the critical flaw size.

By assuming that the fracture toughness of the fibers remains constant and that the characteristic fiber strength, σ_0, is related to the mean oxide layer thickness according to Eq. (5), i.e., when $a = \alpha$, then the time dependence of the fiber characteristic strength will be given by

$$\sigma_0(t) = \overline{\sigma}_0, \quad t \le \frac{1}{k}\left(\frac{K_{\mathrm{IC}}}{Y\overline{\sigma}_0}\right)^4 \tag{6a}$$

$$\sigma_0(t) = \frac{K_{\mathrm{IC}}}{Y\sqrt[4]{kt}}, \quad t > \frac{1}{k}\left(\frac{K_{\mathrm{IC}}}{Y\overline{\sigma}_0}\right)^4 \tag{6b}$$

Note that Eqs. (6) indicate the existence of an incubation period equal to the time required to grow an oxide layer as thick as the size of the average critical flaw in the virgin fibers.[§] Afterward, the characteristic fiber strength changes with time as $\approx t^{-1/4}$.

To predict the time-to-failure and the elongation of a fiber bundle subjected to a constant load at a temperature at which the fibers oxidize but do not creep, we make use of Eqs. (1) and (2), which form an iterated function system (IFS). The solution to the IFS at each time step will provide the evolution of the probability of failure of the fiber bundle and, through Eq. (3), its elongation. Failure of the bundle is associated with the divergence of the IFS. Additional details about this analysis can be found elsewhere.[7,13]

Using the values listed in Table I, numerical simulations of the stress-rupture of fiber bundles of CG-Nicalon fibers in ambient air at 950°C were conducted. Figure 5 shows the predicted strain histories for fiber bundles subjected to constant initial bundle stresses of 400, 500, and 600 MPa. These stress levels were chosen considering that in the 2-D composite only one half of the fibers are aligned with the loading direction. Therefore, in a composite with a fiber volume fraction of 40%, the stress acting on the fibers bridging the plane of the critical transverse matrix crack will be initially 5 times the composite stress.

Note that although the overall fiber bundle stress is kept constant (F/AN_0) in the analysis, the fiber stress is not constant (F/AN) and the latter increases as the number of failed fibers increases.

Except for a short incubation time period that is imperceptible in Fig. 5, the strain versus time curves have positive concavity in agreement with the curves recorded during the stress-rupture evaluation of the CG-Nicalon/enhanced SiC CFCC (Fig. 2). Note also that the strain versus time curves in Fig. 5 show accelerated deformation prior to failure.

Although others have referred to the deformation history curves recorded during the stress-rupture testing of these materials as ''creep curves,''[8] it should be pointed out that the time-dependent deformation of these materials during stress-rupture testing at temperatures up to 1000°C is not in reality the result of creep (i.e., flow). Rather, the increasing compliance of these materials with time results from the progressive breakdown of the fiber bundles in the composite, as demonstrated here.

Figure 6 shows the time evolution of the probability of failure associated with the model predictions presented in Fig. 5, according to Eq. (1). The results in Fig. 6 indicate that it only takes the failure of about 20% of the fibers in the bundle to induce the failure of the entire fiber bundle.

The predicted time-to-failure for the fiber bundles has been plotted in Fig. 4. As expected, the life of the fiber bundles decreases with increasing bundle stress. Note that the slope of the rupture-stress versus time-to-failure curve for the fiber bundles has a slope value of −1/4, which is consistent with Eq. (6), and that such a slope value is comparable to that of the curve that describes the experimental results for the CG-Nicalon/enhanced SiC CFCC. Note again that the initial stress levels used for the bundle life predictions were selected based on the fiber volume fraction in the composite (i.e., one half of 40%) and therefore are 5 times larger than the composite stresses. The predicted times-to-failure for the fiber bundles subjected to stress-rupture conditions are comparable to the experimental values obtained for the composite for each stress level (Fig. 4). This agreement was achieved by using a gauge

Table I. Numerical Values Used in Model Predictions

E	200 GPA (Ref. 17)
ℓ	0.0035 m
ℓ_0	0.025 m (Ref. 17)
m	3.8 (Ref. 17)
σ_0	1.8 GPa (Ref. 17)
k	exp(11.34 − 8887/T) nm^2/min (Ref. 15)
K_{IC}	0.8 MPa·m$^{0.5}$

[†]It is assumed that the oxide scales do not bridge between fibers.

[‡]Although in a cylindrical geometry the surface through which molecules diffuse is not constant, experimental results have demonstrated that the classical parabolic law derived for planar geometries fits the oxidation data for fibers very well.[15,16]

[§]It is found that at short oxidation times the strength of SiC increases because the oxide layer heals surface defects. However, oxidation results in strength reduction at longer times. Here it is assumed that the characteristic strength of the fibers remains constant until the mean oxide layer thickness becomes larger than the critical defect size associated with the characteristic strength of the original virgin fiber strength distribution.

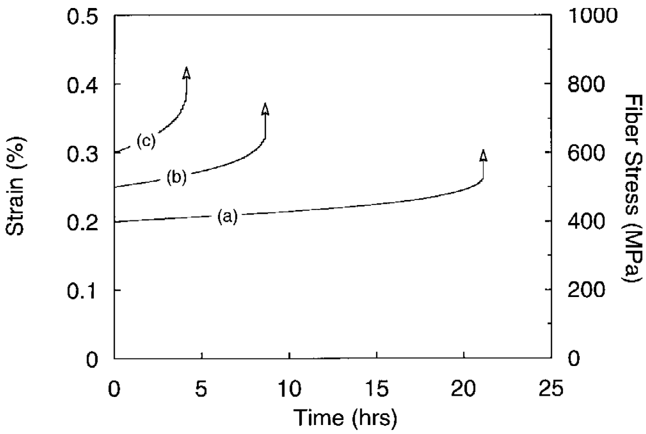

Fig. 5. Predicted strain versus time curves for Nicalon fiber bundles subjected to constant tensile load in ambient air at 950°C. Initial bundle stresses (a) 400, (b) 500, and (c) 600 MPa. Also shown is the fiber stress. Data in Table I were used for model predictions.

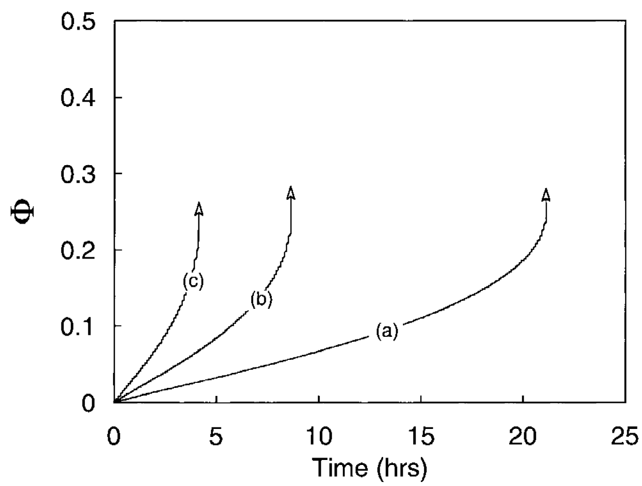

Fig. 6. Predicted time evolution of the probability of failure for Nicalon fiber bundles subjected to constant tensile load in ambient air at 950°C. Initial bundle stresses (a) 400, (b) 500, and (c) 600 MPa.

length value of 3.5 mm in the simulations. Although this gauge length value is comparable with the wavelength of the fabric ≈3 mm, it is likely that the effective gauge length of the reinforcing fiber bundles is even shorter because, due to the waviness of the fiber fabric, which was not considered in the analysis of the fiber bundles, the stress acting on the fibers will be higher as a result of bending.

Although the analysis presented above is very simple, it predicts rather well the trend of the experimental stress-rupture behavior of CG-Nicalon/enhanced SiC CFCCs in ambient air at 950°C. This agreement is remarkable considering that the analysis neglected details such as the spacing of matrix cracks, the oxidation of the fiber coating, the effect of fiber architecture, and the nonuniform stress distribution acting on the fibers, etc., and that the only adjustable parameter in the numerical simulations was the gauge length of the fiber bundles.

In the formulation of the analysis it was assumed that the oxide layer on the fibers, which controlled their strength, grew uniformly with time. In the composite, however, because of the presence of matrix cracks, the oxidation of the fiber coating, and the sealing of the resulting channels between the fibers and the matrix, the thickness of the oxide layer will not be uniform. Therefore, a more realistic analysis should account for the nonuniformity of the oxidation of the fibers, but at the expense of added complexity.[7]

It appears that the critical steps leading to the rupture of this material are the cracking of the matrix, the ingress of the environment to the interior of the material, and the oxidation of the fibers. Furthermore, the combination of the experimental results and the analysis presented above suggest that for the range of stresses considered in this investigation, the oxidation of the fiber bundles is the dominant mechanism that controls the life of the composite. However, it is expected that the micromechanical and thermochemical details that were neglected in this analysis will play a more important role at lower stresses near the matrix cracking stress.[6,7] For example, recent experiments have shown that comparable CFCCs reinforced with 8-harness satin weave fabric exhibit a larger endurance limit than comparable materials reinforced with plain weave fabric.[18]

V. Conclusion

It was found that under stress-rupture testing conditions in ambient air at 950°C in air, CG-Nicalon/enhanced SiC CFCCs exhibit delayed failure, that the compliance of the material increases with time, and that the material exhibits accelerated deformation prior to failure. It was shown that a very simple model based on the oxidation-induced stress-rupture of fiber bundles predicts a time dependence for the loss of strength of fiber bundle of the form $t^{-1/4}$, which was in good agreement with the experimental results for the composite. It was also demonstrated that such a model can account for the increasing compliance recorded during the stress-rupture tests, and accelerated deformation prior to failure. The critical steps resulting in the rupture of this material are the cracking of the matrix, the ingress of the environment to the interior of the material, and the oxidation of the fibers, and for the stresses investigated the oxidation-induced stress-rupture of the fibers appears to be the dominating mechanism.

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