Tensile strength and creep behavior of carbon-carbon composites at elevated temperatures

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Abstract

Tensile strength and creep behavior of a 2D laminate carbon-carbon composite (C/C) were examined from room temperature to 2773 K in an inert atmosphere. The tensile strength of the C/C was monotonically enhanced with increasing test temperatures. In particular, significant improvement was observed at temperatures higher than 1873 K. In this temperature range, nonlinear stress-strain curves were observed at low deformation rates. The source of the apparent nonlinearity was concluded to be creep deformation. Two ruling mechanisms for the strength enhancement of the C/C at elevated temperatures were identified. The first source was de-gassing of absorbed water. The second was creep deformation. This phenomenon was notable at temperatures higher than 1873 K, and produced much larger enhancement than the de-gassing.

1 Introduction

Carbon-Carbon composites (C/Cs) are the only light weight material that can maintain high strength and toughness at temperatures higher than 2273 K. In light of this strong advantage, C/Cs are now in use in various high temperature structures, e.g., heat resistant components in rocket nozzles and thermal protection structures in space vehicles [1-4]. However, the mechanical properties of C/Cs at elevated temperatures are not well understood. Only a few reports have been published concerning this issue [3-6], but the results have been contradictory even in regard to the temperature dependence of tensile or flexural strength. These confusing results might come from the lack of systematic research on the fracture behavior of C/Cs at elevated temperatures. For polycrystalline graphite, tensile strength at room temperature was enhanced by de-gas treatment up to the strength level at ≈ 1773 K [7], and creep deformation was clearly observed at temperatures higher than 2273 K [8]. The temperature dependence of strength and the stress-strain (σ - ε) relation is essentially unknown at temperatures higher than 2273 K. In this temperature range, C/Cs have been reported to exhibit creep deformation [9].

In the present study, tensile tests of a 2D laminate C/C parallel to the fiber axis were conducted at various temperatures up to 2773 K. The ruling mechanisms for the strength enhancement of the C/C at high temperatures were discussed in terms of the effects of constituent materials and fiber-matrix interfacial strength. Temperature dependences of the matrix was experimentally evaluated by 45° off-axis tensile tests, and bonding strength along the fiber/matrix interface was estimated from the transverse tensile strength of an unidirectionally reinforced C/C fabricated by the same procedure as the 2D laminate C/C. The effects of thermal stress and the de-gas effect on tensile strength are also discussed based on the experimental and simple stress analysis of fiber and matrix.

2 Experimental procedure

2.1 Materials

The C/C used in this study was supplied by Across Corp., Japan, and was fabricated using the pre-formed yarn method [10]. PAN-based 6000 filament carbon fiber bundle (TORAYCA® M40, Toray, Japan) was used for the reinforcement, and the fiber volume fraction was set to 0.4. A symmetric cross-ply C/C (CP) with stacking sequence [0/90]4s was used to evaluate the mechanical properties of the C/C by loading parallel to the fiber axis. Unidirectional C/Cs (UDs) were additionally used to roughly estimate

the bonding strength along the fiber-matrix interface.

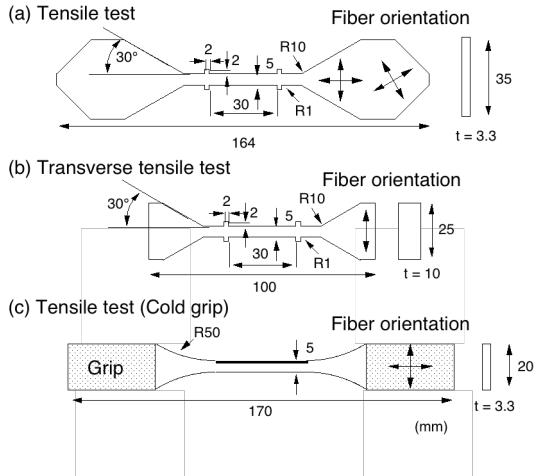

Fig. 1. Shape and dimensions of specimens, (a) tensile and 45° off-axis tensile test, (b) transverse tensile test of the UD and (c) tensile test using cold grip.

2.2 Tensile tests

Tensile tests were carried out using the three types of specimen geometry shown in Figures 1 (a) - (c). Specimen (a) was used for tensile tests of the CP in the fiber axis (0°) and 45° off-axis directions. The off-axis tests were performed for the evaluation of shear behavior. Tensile tests in the transverse direction (90°) were also conducted using the specimen shown in Figure 1 (b). In the tensile specimens of Figures 1 (a) and (b), the overhung sections supported the tensile load. To estimate the effect of stress concentration due to the overhang, 0° tensile tests of CP using the specimens shown in Figure 1 (c) were conducted using water-cooled hydraulic gripping. For these specimens, copper tabs were fixed to the gripping areas in order to avoid fracture around the gripping areas.

Tensile tests of specimens (a) and (b) were carried out using a screw-driven universal testing machine (AG-10G, Shimadzu Corp., Japan) equipped with a furnace having a high temperature capability up to 3273 K in an inert atmosphere. Strain during the tensile tests at elevated temperatures was determined using an extensometer directly attached to the gage area of a specimen (Model 632.59, MTS systems Co., USA). The specimens were loaded via a polycrystalline graphite test fixture. In the tests, a whole specimen was set in a uniform heating zone of the furnace. The temperature of the specimen was measured using a thermocouple (Type C) set near the gage section of the specimen up to the temperatures of 1873 K and using an optical pyrometer at temperatures higher than 1873 K.

To confirm the effect of stress concentrations due to the overhang, tensile tests at 2273 K were also conducted using another screw-driven testing machine (Model 808, MTS systems Co., USA) having water-cooled hydraulic gripping. In this facility, only the gage section of the tensile specimen (40 mm in length) was heated and strain was measured by means of an extensometer directly attached to the gage area of a specimen (Model 632.59, MTS systems Co., USA).

In all the tests, a specimen was heated with a constant heating rate of 10 K/min up to test temperature. After maintaining the temperature 30 min, tensile tests were initiated. Most of the tensile tests were carried out at a constant crosshead speed of 0.1 mm/min. To confirm the time-dependent effect on mechanical properties, the crosshead speed was occasionally varied up to 5.0 mm/min.

The tensile strength of graphite at room temperature was known to be enhanced by de-gas treatment [7]. In order to confirm this effect in C/Cs, part of the CP specimens underwent de-gassing treatment using the following procedure under vacuum before tensile tests; A sample was 1) heated at a constant heating rate of 10 K/min to 1573 K, 2) held at 1573 K for 1 hour for the evolution of absorbed gases, and 3) cooled to room temperature at the same rate and left for 1 hour at room temperature.

2.3 Creep tests

In order to understand creep behavior, creep tests were conducted using the tensile testing machine described formerly (AG-10G, Shimadzu Corp., Japan) in an inert atmosphere under a constant load. The specimens in this test were heated up to test temperatures by a manner identical to the tensile tests and then loaded to desired stress levels at a constant loading rate of 0.9 KN/min. The creep tests were performed under various stresses and temperatures in order to estimate the parameters in the Dorn equation expressed in terms of stress exponent, n, and activation energy, Q.

3 Result and Discussions

3.1 Tensile behavior in the fiber axis direction

Figure 2 shows typical stress-strain, σ - ε, curves of the CP loaded in the 0° direction at room temperature and 2273 K at crosshead speeds of 0.1 and 5.0 mm/min. The stress-strain curves slightly swayed because of the electric noise from the extensometer.

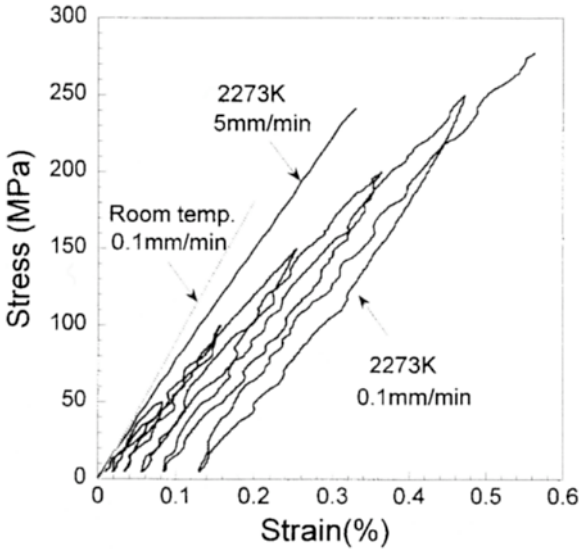

Fig. 2. Stress-strain curves of the CP at room temperature and at 2273 K with different crosshead speed of 0.1 mm/min and 5.0 mm/min.

As shown in this figure, the σ - ε curve at room temperature was linear up to the total fracture and traced the same line during repetition of loading-unloading. In contrast at 2273 K, the σ - ε curve showed significant nonlinear deformation accompanied by non-recovery strain at test speed of 0.1 mm/min. From the σ - ε curves, Young's modulus of the CP at room temperature was calculated to be 80 ± 3 GPa. This value agrees well with an expected value (78 GPa) estimated from Young's modulus of the fiber (390 GPa [11]) using the rule of mixture without contribution of the matrix [12]. At 2273 K, Young's modulus of the CP estimated from the initial slope in Figure 2 was almost the same as that in room temperature. However, it immediately degraded with applied stress. The tensile strength of the CP was monotonically enhanced with increase in temperature, as shown in Figure 3. This enhancement became rapid from 1773 K. SEM photographs of the fracture surface of the CP after tensile tests indicate no significant difference in the fiber's fracture pattern and pull-out length of the fibers between specimens tested at low and high temperatures.

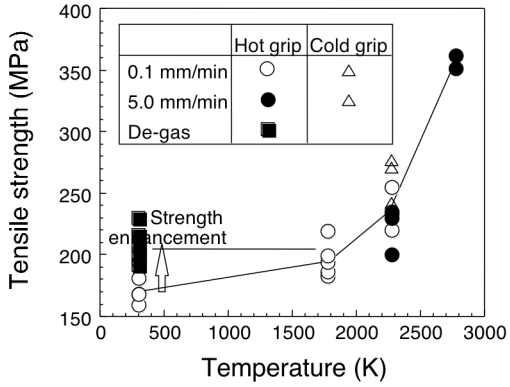

Fig. 3. Tensile strength of the CP as a function of test temperature. Strength enhancement by de-gas are also shown.

3.2 Nonlinear deformation

Strong nonlinear deformation and permanent strain were observed at 2273 K under a test speed of 0.1 mm/min. To identify the mechanisms yielding this nonlinear behavior, the σ - ε curve was also determined at an increased testing speed of 5.0 mm/min at 2273 K. Figure 2 also compares σ - ε relations under test speeds of 0.1 and 5.0 mm/min at 2273 K. As shown in this figure, the σ - ε curve at the test speed of 5.0 mm/min resulted in a straight line up to the total fracture with a slight decrease in Young's modulus compared with that at room temperature shown as a broken line in Figure 2. This fact suggests that, at elevated temperatures, the σ - ε curves of C/Cs are still linear and apparent nonlinear behavior appears due to a strain rate-dependent factor, probably creep deformation.

Figure 4 shows creep strain vs. time curves for the CP obtained under constant stresses. As is similar to the usual creep deformation behavior, the first stage and the second stage, steady-state creep, are clearly recognized in Figure 4. From these results, Figures 5 (a) and (b) were obtained for the determination of the activation energy, Q, and the stress exponent, n, in eq. (1), respectively. Thus, Q and n were estimated as 730 KJ/mol and 2.0. In these figures, reported data [16] were also plotted. These data were generated using a similar C/C, supplied by the same supplier (Across Corp.) and fabricated by the same procedure (preformed yarn method) as the present study, but using different

fiber (TORAYCA® T-300, Toray Corp.). It follows from Figures 5 (a) and (b) that the Q value of these two C/Cs appears identical if the data at 1873 K is neglected, but n is different. The Q value is smaller than that reported for other C/Cs [9] and glassy carbons [14], 1000 to 1200 KJ/mol. Regarding n, 6.0 [13] and 8.0 values have been reported for other carbon materials [9, 14]. The difference in fiber might bring different n value and the same matrix bring similar Q values, however, the origin of this phenomenon has not been elucidated yet. Finally the C/C used in this study exhibit creep deformation from 1873 K. Thus, the creep deformation is established to be a source of non-linear behavior on the strain-stress curve under low strain rate.

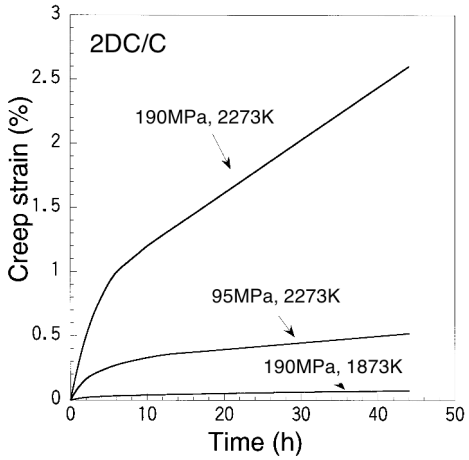

Fig. 4. Typical examples of time vs. strain curves of the CP.

3.3 Temperature dependence of tensile strength

As shown in Figure 3, the tensile strength of C/C was enhanced with increasing temperature. However, controlling mechanisms for this enhancement have not been clarified. Six possible mechanisms are discussed in the following.

(A) Effect of absorbed gas

The evolution of absorbed substances, probably water, has been reported to enhance the tensile strength of polycrystalline graphite [7]. Therefore, similar enhancement can be expected in the tensile strength of the present C/C. In Figure 3, tensile strength after de-gas treatment is compared to that without treatment at room temperature. This comparison clearly manifests that tensile strength increased to about 205 MPa, which is equal to tensile strength at 1873 K. Thus, the de-gas phenomenon was the primary mechanism responsible for strength enhancement of the C/C up to ≈ 1873 K.

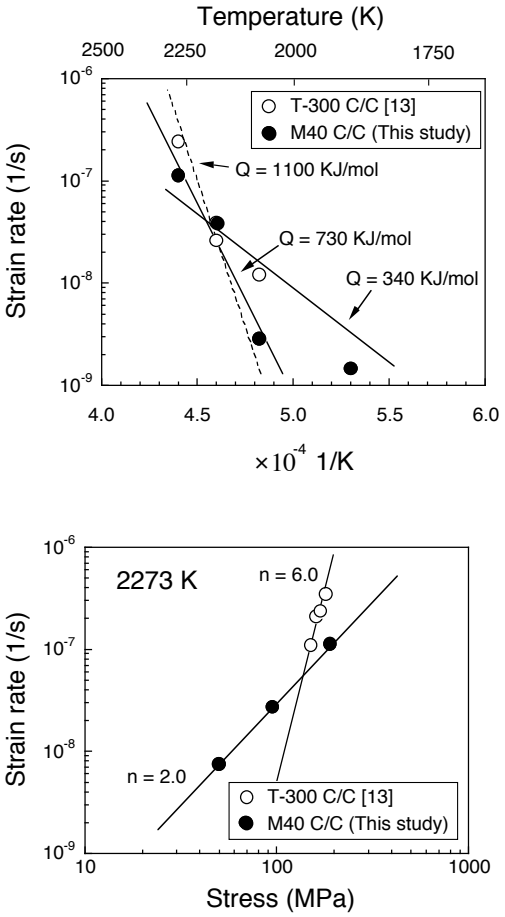

Fig. 5. Steady state creep rate of the CP showing (a) activation energy between 1773 K and 2273 K and (b) stress exponent at 2273 K. The values from the reference are also shown in the figures.

(B) Dependence of matrix strength on temperature

Many researchers presume that the ultimate tensile fracture of C/Cs is dictated by the fracture strain of the matrix [12, 15-17]. Thus, the matrix strength was estimated as a function of test temperature by 45° off-axis tensile tests of the CP. Matrix strength enhanced with temperature with almost same tendency as the strength of the C/C up to temperature of 2273 K. This fact implies that matrix strength is one of the ruling factors for the enhancement of the C/C strength at elevated temperatures.

(C) Fiber-matrix interface

The Bonding strength of the fiber-matrix interface is an important factor for controlling the

tensile fracture of brittle matrix composites. The tensile strength of C/Cs is generally enhanced by decreasing the bonding strength along the fiber-matrix interface [15-17]. In the present study, the fiber-matrix interface bonding strength was estimated by transverse directional tensile tests using the UD specimen. The transverse tensile strengths of the UD at room temperature and 2273 K were measured as 3.0 ± 0.1 MPa and 4.3 ± 0.5 MPa, respectively. These results imply that the interfacial bonding strength at 2273 K is higher than that at room temperature. From the viewpoint of low interfacial strength yielding high tensile elongation [15-17], it can be understood that the strength enhancement of the C/C at elevated temperature demonstrated in Figure 3 cannot be explained in relation with the fiber-matrix interface bonding strength.

(D) Effect of creep deformation

As shown in Figure 6, the tensile strength of the CP at 2273 K was enhanced with decreasing test speed from 5.0 mm/min to 0.1 mm/min. In addition, the tensile strength after creep test was enhanced further. Thus, the similar phenomenon is expected to occur in matrix and fiber during creep deformation of the C/Cs and contributes to strength enhancement of the C/C at temperatures > 1873 K.

(E) Effect of thermal stress

Due to the mismatch in the coefficient of thermal expansion (CTE) between the fiber and matrix, thermal stresses should be induced in C/Cs after processing. The thermal stresses are gradually released with temperature increase. Thus, the thermal stress might affect tensile strength. From the CTEs of the fiber and matrix as shown in Table 1 [5], tensile stress on the fiber-matrix interface was estimated at room temperature. Since tensile interface stress is expected to weaken the bonding strength of the interface, this result indicates that the bonding strength of fiber-matrix interface in the C/C was enhanced with the release of thermal stress at elevated temperatures. On the contrary, the apparent tensile strength of the matrix in the C/C should be enhanced with temperature by the release of tensile thermal stress in the matrices. However, as already discussed, de-gassing had sufficient effect to explain the tensile strength enhancement of the C/C up to 1873 K at room temperature. Hence, thermal stress in the matrices by processing was supposed to be almost completely released at room temperature.

Table 1 Coefficient of thermal expansion of the fiber and matrix

		Fiber	Matrix
Coefficient of thermal expansion ($\times 10^{-6}$ 1/K)	⫽	-1.3 ~ 1.5	3.9 ~ 6.9
	⊥	10	

⫽ Parallel to the fiber axis
⊥ Perpendicular to the fiber axis

(F) Fiber strength

Only a few studies have focused on the tensile strength of carbon fiber at elevated temperatures [18-21]. Recently, Sauder et. al [21] conducted tensile tests on PAN based carbon fiber heat treated 2473 K and showed that the tensile strength of the carbon fiber slightly increased from room temperature to 2273 K. This strength enhancement of PAN based carbon fiber was explained to result from a reduction in flaw severity associated with internal stress relaxation [21]. This fact shows no significant variation in tensile strength expected at the temperatures below HTT. The HTT of the fiber in the present study, M-40, is over 2500 K. Thus, a tendency similar to the result of Sauder et. al can be assumed for M-40. On the other hand, the fracture strain of the fiber increase with temperature. Since, Young's modulus of the fiber degrades from 1273 K and significantly degrades > 1873 K with slight increase in tensile strength [18-21]. This fact means that the fiber supposed to become tougher at elevated temperature from breaking by crack or defect in the matrix. This fracture strain enhancement of the fiber can be another source of slight strength enhancement of the C/C.

Conclusions

The temperature dependence of the tensile strength and deformation of a laminated C/C were examined up to 2773 K. In order to explain the high temperature behavior of the C/C, the effects of various factors were discussed. These factors included the high temperature properties of fiber, matrix, and the fiber-matrix interface, the effects of the evolution of absorbed gas, creep deformation, and thermal stress. The conclusions are as follows:

1. The tensile strength of the C/C was enhanced with temperature.
2. Two mechanisms were found to be responsible for the strength enhancement of the C/C at elevated temperatures. (1) Evolution of absorbed gas induced an improvement in matrix strength. This mechanism

was effective up to 1773 K. (2) creep deformation at temperatures higher than 1773 K.
3. The stress-strain curve of the C/C was linear up to the total fracture, at least up to 2273 K without creep deformation.
4. Creep deformation in the C/C was observed above 1873 K. Activation energy, Q, and stress exponent, n, in the temperature range between 1873 K and 2273 K were 730 KJ/mol and 2.0, respectively.

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