Tension-Tension Fatigue of 2.5D-SiC/SiC Ceramic Matrix Composites at Elevated Temperatures

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Abstract. In order to investigate the influence of Tension-tension fatigue behavior of fiber reinforced ceramic-matrix composites, SiC/SiC composites with 2.5D fiber architecture were prepared via Precursor Infiltration and Pyrolysis (PIP) method. Tension-tension fatigue behavior of SiC/SiC composites was investigated in 1000°C and 1200°C, under fatigue stresses ranging from 70 to 120MPa at a frequency of 1.0 Hz, under oxidizing atmosphere. Fatigue life of SiC/SiC composites achieved 305,984 cycles at 1200°C/70MPa, and 252,988 cycles at 1200°C/108MPa, which were longer than test in 1000°C. Composite microstructure, as well as failure mechanism was investigated. The main reason that cause tension-tension fatigue failure of SiC/SiC composites are oxidation damage and stress damage. The microstructure of SiC/SiC composites demonstrated that the oxidation of interfaces and SiC fibers caused the failure of the composites. Under the load of alternating stress, the original voids and cracks in the material preparation process will be widened, and new cracks will be generated, for cracked SiC/SiC composites, oxygen in is especially serious at intermediate temperatures (~800°C) where it can reach the SiC fiber before being sealed off by slow-glowing silica on the matrix crack surface, therefore, tension-tension fatigue life of SiC/SiC composites at 1000°C is lower than 1200°C.

1. Introduction
Continuous fiber reinforced silicon carbide ceramic matrix composites have the advantages of high temperature resistance, high strength and stiffness and low density. Because of fiber-reinforcement, the SiC/SiC composites are more damage tolerant and have the capability for larger components than their SiC monolithic counterparts, and is expected to replace high-temperature alloys and refractory metals as high-temperature structural materials[1-3]. New materials for engineering applications must solve the problem of characterization of mechanical behavior. Studying the failure mechanism of materials helps to improve the process and improve its performance. In recent years, the tensile and compressive properties, shear properties, fatigue properties and fracture toughness of woven SiC/SiC ceramic matrix composites under normal temperature conditions have been reported at home and abroad [4–7].

However, the above research content is only the regular general mechanical properties, and does not involve its mechanical properties in the service environment. Ceramic matrix composites are
widely used in high temperature structural components such as heat exchangers in aerospace applications, aerospace engine nozzles, and rocket engine thrust chambers. These high temperature structural components are mostly subjected to high temperature cyclic loading during service. For the thermal structural components of aero-engines, the composite is subjected to an oxidizing gas at a temperature of 1200 °C, and the internal temperature of the engine is as high as 1450 °C or more. Based on the requirements of high temperature performance and overall weight reduction, continuous silicon carbide fiber toughened silicon carbide ceramic matrix composite (SiC/SiC) has become one of the main candidates for the new generation of aerospace engine thermal structural components with its good overall performance.

When used as a structural material, its fatigue behavior must be considered. It has been shown that the properties of SiC/SiC composites decrease with the increase of fatigue cycles, the elastic modulus decreases significantly, and the cumulative strain and residual strength increase. These macroscopic phenomena are the result of various damages inside the material during fatigue process. The damage forms mainly include delamination, fiber bundle bifurcation, matrix cracking, fiber breakage, fiber bridging, and interface debonding[8-11].

Ojard [12] studied the effects of test frequency and environmental factors on the fatigue properties of SiC/SiC composites. The test conditions were 1200 °C in air or water vapor atmosphere, the frequency was 0.1, 1.0, 10 Hz. The fatigue limit and fatigue life of the material decrease with the increase of the frequency. The water vapor greatly deteriorates the fatigue properties of SiC/SiC composites. The room temperature retention performance after fatigue test in cold air shows that the tensile strength does not decrease and the elastic modulus decreases up to 22%, indicating that the fiber was not damaged in the fatigue test, and the matrix produced cracks.

Morscher [13] studied the strength retention, damage evolution and failure mechanism of SiC/SiC composites prepared by MI process after high temperature tensile fatigue test. The results show that tensile strength and elastic modulus at room temperature increase with increasing stress and time. The modulus retention rate continues to decrease. The failure mechanism of performance degradation under different test conditions is different: under short-term high stress, the oxidation-induced non-bridge chain matrix crack growth causes the load redistribution in the intact region of the composite material and the local stress concentration at the crack tip, eventually with time one of these cracks develops into a crack; in the long-term low stress, the inherent creep control of the fiber reduces the growth mechanism or the free silicon penetrates the CVI-SiC interface layer to erode the fiber, resulting in a decrease in fiber strength.

The damage evolution behavior of the SiC/SiC composites under fatigue loading is very important for the design of high temperature parts. It has been shown that the fiber weaving method, the material preparation process and the fatigue test parameters all have influence on fatigue behavior and damage form of SiC/SiC composites. In this study, the tension- tension fatigue properties of 2.5D SiC/SiC composites prepared by PIP process at 1000 °C and 1200°C were studied. The fracture damage mechanism of composites was studied in combination with fracture morphology.

2. Materials and Test Specimens

2.1. Test Material
The material studied in this research effort was manufactured by AVIC Aviation foundation technology establishment, by Precursor Infiltration and Pyrolysis (PIP) of SiC into the 2.5D woven KD-II SiC fiber performs. The composite was supplied in the form of 3.5mm thick plates. The composite consisted of 15 plies of KD-II [0°/90°] fabric woven in an 2.5D woven performs. Prior to matrix densification, the performs were coated with carbon (PyC) layer in order to decrease interface bonding between fibers and matrix, thereby increasing composites strength and toughness. The thickness of fiber coating was ~250nm. The SiC matrix was densified by PIP process, the precursor was provided by SAIFEI co., LTD., the softening point of solid polycarbonsilane was 180–200 °C, and solid polycarbosilane and xylene were prepared into solid polycarbosilane precursor solution with a mass ratio of 1:1. The viscosity of the precursor solution at 25°C was (20–40) mPa•s.
The volume fraction of the fibers was 46% and the porosity of the composite was 12.9%. The density of the composite was 2.38g/cm³. The tension-tension fatigue and tensile strength specimens had an outer seal coating of SiC that was applied by chemical vapor deposition (CVD) after the specimens had been machined.

2.2. Preparation of interface layer
PyC interface layer of SiC fiber was applied by chemical vapor infiltration (CVI). The deposition were carried out in argon flow of 200ml/min. Deposition temperature were 950°C, deposition pressure were 5KPa, and deposition time 20h.

2.3. Preparation of SiC/SiC composites
SiC/SiC composites were prepared by PIP process until the weight gain rate decreased to less than 2%. The density of SiC/SiC composites was 2.34 g/cm³ and the porosity was 8.26%. In order to ensure the comparability of the prepared composite materials, the PIP process conditions of the three materials are exactly the same. Process flow of SiC/SiC composite prepared by PIP process was in Figure 1.

![Figure 1. Process flow of SiC/SiC composite prepared by PIP process.](image)

2.4. Test Specimen Preparation
SiC outer seal coating of SiC/SiC composites was applied by CVD method, and the coating thickness was about 200nm. Deposition temperature were 1000°C, deposition pressure were 5KPa, and deposition time 20h. The prepared SiC/SiC composite material is machined to 3.5mm thick plates. Its external dimensions are shown in Figure 2. SiC/SiC composite plates are cut along the longitudinal direction with diamond cutters into test pieces in the form of Figure 2.

![Figure 2. Schematic diagram of SiC/SiC composite material external dimensions.](image)

3. Experimental Arrangements and Procedures

3.1. Test Procedures

3.1.1. Monotonic Tensile Test. High temperature tensile strength test was conducted by Chinese building materials inspection certification group co., LTD., test methods refer to GB/T 36264-2008 "Test method for tensile strength of fiber composites under ultrahigh temperature and oxidizing
environment". High temperature tensile strength tests were conducted in laboratory air. The specimen was heated at a rate of 15°C/min to test temperature, then the specimens tested in air were held at test temperature for 10 minutes. During the test, the loading speed was 1.2mm/min, until the sample was destroyed, and the load-time curve was recorded. Tensile strength is calculated according to equation (1):

$$\sigma_t = \frac{P_{\max}}{b_1 \times h}$$

(1)

$\sigma_t$ —— Tensile strength, MPa;
$P_{\max}$ —— Maximum load, N;
$b_1$ —— Sample width, mm;
$h$ —— Sample thickness, mm.

3.1.2. Tension-Tension Fatigue Tests. High temperature fatigue test was conducted on Instron8801 hydraulic servo fatigue test machine. Test methods refer to Q/ZHFC 8533-2018 "Test method for tensile fatigue properties of fiber reinforced ceramic matrix composites under high temperature oxidation environment". The test furnace and installation schematic diagram are shown in Figure 3. Test specimen were mounted into the 10t machine via Instron8801 hydraulic, water cooled, wedge grips.

Tension-tension fatigue tests with an R ratio (ratio of minimum stress to maximum stress) of 0.1 were conducted in load control at frequencies of 1Hz in laboratory air, the sinusoidal wave is selected. Fatigue run-out was defined as $10^6$ cycles. The peak stress was 70MPa, 108MPa and 120MPa. During the loading process, the load-displacement curve was recorded at the same time. In all tests, the specimen was heated under a zero load command at a rate of 15°C/min to test temperature, then the specimens tested in air were held at test temperature for 10 minutes. The fatigue stress is calculated according to equation (1.4.2):

$$S = \frac{P}{b \times h}$$

(2)

$S$ —— Fatigue stress, MPa;
$P$ —— Fatigue load, N;
$b$ —— Sample width, mm;
$h$ —— Sample thickness, mm.

![Figure 3. Schematic diagram of test furnace and installation.](image-url)
3.2. Microstructural Characterization
After testing, the failed samples were examined under S-4800 scanning electron microscope Produced by HITACHI, Japan.

4. Results and Discussion
4.1. S-N curve of SiC/SiC composites
Before the tension-tension fatigue test, unidirectional tensile tests were carried out on samples of the same batch at 1000°C and 1200°C, and the results showed that the tensile strength of SiC/SiC composites was 208MPa at 1000°C and 200MPa at 1200°C. According to the static tensile strength of SiC/SiC composites, the peak stress of fatigue test was determined to be 35%, 55% and 60% of the tensile strength, 70 MPa, 108 MPa and 120 MPa respectively. Maximum fatigue stress-life curves of 2.5D-SiC/SiC composites at 1000°C and 1200°C are shown in Figure 4.

![Figure 4](image)

**Figure 4.** Figure stress-life curves of 2.5D-SiC/SiC composites at 1000°C and 1200°C.

Existing studies[14] have shown that, the fatigue life of SiC/SiC composites can be roughly divided into three areas: short life area (I area, N<10⁴ cycles), medium life area (II area,10⁴<N<10⁶ cycles), and long life area (III area, N>10⁶ cycles). The fracture occurred in the low life area when the stress approached to the tensile strength. When the stress is between fracture strength and fatigue limit, the fracture occurs in the medium life zone. When the stress is less than the fatigue limit, the fracture occurs in the long life zone. As can be seen in Figure. 4, when the maximum fatigue stress is between 115MPa and 120MPa, most of the fractures occur in the short life area, and when the maximum fatigue stress is between 70MPa and 115MPa, most of the fractures occur in the middle life zone. When the maximum fatigue stress rises from 70MPa to 108MPa, the failure cycle number increases less, and the decline slope of the maximum fatigue stress-life curves is slow. However, when the maximum fatigue stress rises from 108MPa to 120MPa, the maximum fatigue stress-life curves has a urgent decline slope, at which time the life of composite material decreases sharply. By comparing the S-N curves at 1000°C and 1200°C, it can be seen that the S-N curve of SiC/SiC composite material at 1200°C is above the curve at 1000°C, indicating that under the same maximum fatigue stress, the life of the material at 1200°C is longer than that at 1000°C, while the tensile strength at 1000°C is slightly higher than that at 1200°C, which may be caused by the severe oxidation of the material at 1000°C.
4.2. Microstructure of the Specimens tested in Tension to fatigue at different temperature

Figure 5 shows the fatigue fracture of 305,984 cycles in an air environment of 1200°C. It can be seen that the fracture of the sample shows ductile fracture characteristics, but the fiber extraction length is short. Toughening mechanisms such as crack deflection and fiber bridge chain can be seen in SiC matrix, indicating that the failure mechanism of materials under tensile fatigue load of 1200°C and 70MPa is similar to that of uniaxial tensile strength.

Figure 6 shows the fatigue fracture that circulates 288,001 times in an air environment of 1200°C. It can be seen that the fracture of the sample shows a penetrating brittle fracture characteristic with relatively smooth fracture and no fiber pulling out. As can be seen from the local magnification figure, SiC matrix presents a glassy structure, indicating that the matrix has been oxidized. A gap can be seen on the fiber surface, not only the PyC interface layer cannot be seen around the SiC fibers, but the boundaries between SiC fibers and the matrix in some areas is also very fuzzy, indicating that not only the matrix and PyC interface layer are oxidized, and SiC fibers are also partially oxidized. EDS results show that there are O atoms in both the SiC matrix and the fiber, which proves that the matrix and the fiber have been oxidized, that eventually leading to fatigue failure.

Figure 7(a) shows the fatigue fracture that circulates 252,988 times in an air environment of 1200°C. Figure 7(b) shows the fatigue fracture that circulates 171,010 times in an air environment of 1200°C. It can be seen from Figure 7(a) that the SiC fiber is structurally intact, but there is a gap around the fiber, indicating that the PyC interface layer has been oxidized; as shown in Figure 7(b), the SiC fiber, the PyC interface layer and the SiC matrix have been integrated into one body. The SiC matrix appeared glass morphology obviously. It can be judged that after the fatigue experiment at 1000°C and 1200°C, the samples were oxidized. However, the oxidation of sample fracture was more serious after 1000°C test.
Figure 7(a). SEM micrographs of specimen tested in fatigue at 1Hz in air at (a) 1200°C ($\sigma_{\text{max}}=108\text{MPa}, N_f=252988, R=0.1$) and (b) 1000°C ($\sigma_{\text{max}}=108\text{MPa}, N_f=177010, R=0.1$).

Figure 8. The EDS curves of SiC fibers after tension-tension fatigue test: a. 1200°C ($\sigma_{\text{max}}=70\text{MPa}, N_f=305984, R=0.1$); b. 1000°C ($\sigma_{\text{max}}=70\text{MPa}, N_f=28801, R=0.1$); c. 1200°C ($\sigma_{\text{max}}=108\text{MPa}, N_f=252988, R=0.1$); d. 1000°C ($\sigma_{\text{max}}=108\text{MPa}, N_f=177010, R=0.1$).

4.3. Damage mechanism of tension-tension fatigue of SiC/SiC composites at elevated temperatures

4.3.1 Oxidation damage mechanism of tension-tension Fatigue of SiC/SiC composites at elevated temperatures. Figure 8 shows the original peak curve of EDS analysis of SiC fibers after the tension-tension fatigue test. It can be seen from Figure 8 that no oxygen atoms were detected in SiC fibers after the tension-tension fatigue test at 1200°C/70MPa, indicating that the interface layer can effectively prevent the fiber from being oxidized under this condition. However, oxygen atoms were detected in the fibers of specimens tested at 1200°C/108MPa and 1000°C/70MPa and 108MPa, indicating that the fatigue failure of SiC/SiC composites was caused by oxidative damage under this experimental condition.

4.3.2 Stress damage mechanism of tension-tension Fatigue of SiC/SiC composites at elevated temperatures. Fatigue load have huge effect on the oxidation of materials. Under the load of alternating stress, the original voids and cracks in the material preparation process will be widened,
and new cracks will be generated, which will affect the oxidation behavior of the fibers[15-17]. First, the SiC coating on the surface of the SiC/SiC composite is pulled apart, and the silicon oxide protective film formed on the surface of the coating cannot seal the crack. The oxygen atoms in the air continuously diffuse into the material. Then the SiC matrix is pulled open, the alternating stress causes the crack to penetrate deeper into the material and diffuse through the voids in the SiC/SiC composite to the surface of the carbon phase, reacting with the activation point of the carbon phase. Under continuous stress, the transverse cracks expand forward and deflect the cracks longitudinally. The cracks are bridged by the fibers, so that the cracks continue to expand along the PyC interface layer in the longitudinal direction of the fibers, and the oxygen at the crack tip reaches the transverse cracks of the carbon phase. The bottom part is the starting point to diffuse to both sides of the fiber and oxidize the interface layer and the fiber. After the fiber is oxidized, the expanded profile becomes unclear, and the structure changes to become a weak part in the composite material, which is prone to fracture.

When the stress is small, the crack is hardly opened, and the oxidizing gas mainly relies on the preparation of pores and defects for diffusion, and the speed is very slow, the influence range is small. When the stress is large, the crack is wide open, the concentration of oxidizing gas inside the material is high, and the fiber outside the material is directly oxidized, so the tension-tension fatigue life of the SiC/SiC composite material is drastically reduced when load is increased.

For cracked SiC/SiC composites, oxygen in is especially serious at intermediate temperatures (~800°C) where it can reach the fibers before being sealed off by slow-glowing silica on the matrix crack surface[18-19]. Therefore, it can be explained that the tension-tension fatigue life of SiC/SiC composites at 1000°C is lower than 1200°C. On the other hand, since 1200°C is closer to the material preparation temperature (1150°C~1200°C), the thermal stress of the material can be better released when tested at 1200°C.

5. Conclusion
Through the above research and analysis, the main reason that cause tension-tension fatigue failure of SiC/SiC composites are oxidation damage and stress damage. The reason that fatigue life of SiC/SiC composites at 1200°C is longer than 1000°C, is because (1) 1200°C is closer to the material preparation temperature, so that the thermal stress of materials can be better released at 1200°C. (2) the oxidation damage of SiC/SiC composites was most serious around 800°C, so the oxidation damage was more serious at 1000°C than 1200°C. To improve the fatigue life of SiC/SiC composites, the following two aspects should be considered: (1) prepare dense boron nitride interface layer, to completely protect the fiber and its surface PyC interface, and reduce the oxidation damage of the material. (2) prepare SiC/SiC composite materials with higher density and lower porosity, reduce pores and microcracks in the matrix, thus preventing erosion of oxygen and reducing stress damage.

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