Self-Crack-Healing under Cyclic Stress of Silicon Nitride Composite

Koji Takahashi, Kotoji Ando and Shinji Saito*
Department of Energy and Safety Engineering, Yokohama National University
79-5, Tokiwadai, Hodogaya, Yokohama, 240-8501, Japan
Fax: 81-45-339-4024, e-mail: ktaka@ynu.ac.jp
*NHK Spring Co. Ltd. 3-10 Fukuura, Kanazawa, Yokohama, 236-0004, Japan

Si_3N_4/SiC composite ceramics were hot-pressed and subjected to three-point bending. A semi-elliptical surface crack of 100μm in surface length was made on each specimen. This pre-crack reduced the bending strength by half. The pre-cracked specimens were crack-healed under a cyclic bending stress at 5 Hz, and the resultant bending strength and fatigue strength were examined. The threshold stress for crack-healing, below which the pre-cracked specimens recovered their bending strength, was investigated at healing temperatures between 1273 and 1473 K. The cyclic fatigue strengths of crack-healed specimens were investigated at each healing temperatures. The main conclusions are as follows: (1) A pre-crack can be healed under cyclic stress. The threshold stress for crack-healing was σ_{max}=300MPa at 1273 and 1473 K, which was 75% of the bending strength of the pre-cracked specimens. (2) The crack-healed specimens exhibited quite high cyclic fatigue strength at crack-healing temperatures between 1173 and 1473 K.

Key words: Ceramics, Crack-healing, Silicon nitride, Bending strength

1. INTRODUCTION

Some engineering ceramics possess the ability to heal a crack [1-5]. If this ability is used on structural components in engineering use, great merits can be anticipated, such as increases in the reliability of structural ceramic members, and decreases in the inspection, machining and polishing costs of ceramic components. Si_3N_4/SiC [4,7] and Mullite/SiC [2, 6] with very high self crack-healing ability were developed by the present authors. Some important subjects were investigated using these ceramics, such as (1)optimized crack-healing conditions [4-6], (2)the maximum crack size that can be healed completely [5,6], and (3)cyclic or static fatigue strengths of a crack-healed specimen at elevated temperature [7,8]. Applying optimized crack-healing treatment and subsequent proof testing to the machined ceramic components, the reliability of ceramic components can be increased, simply and economically [9]. Most of the studies on crack-healing were carried out without stress [1-9]

If a crack initiates in service, the reliability will be reduced considerably, because fracture toughness of structural ceramics is not high. If a material can heal a crack in service (under stress and at elevated temperature) and the crack-healed zone has enough strength, it would be desirable for the structural integrity. The present authors have already studied the crack-healing behavior under static or cyclic bending stress, and resultant fatigue strength at the healing temperature for Mullite/SiC [11] and Si_3N_4/SiC [12-14]. It is important to determine the threshold stress for crack-healing, below which a crack can be completely healed. However, the threshold stress for crack healing has not yet been studied. In this paper, we selected Si_3N_4/SiC ceramics having quite a good crack-healing ability as a sample. The following two research objectives were chosen in this study: (1)to investigate the threshold stress for crack-healing, below which pre-cracked specimens recovered their bending strength, at healing temperatures of 1273 and 1473 K. (2)to investigate the monotonic bending strength and cyclic fatigue strength of specimens which crack-healed under cyclic stress at healing temperatures between 1173 and 1473 K.

2. EXPERIMENTAL METHOD

2.1 Material

The silicon nitride powder (SN-E10, Ube Industries Ltd., Ube, Japan) used in this study has the following properties: mean particle size = 0.2μm, the volume ratio of α-Si_3N_4 is about 95%, the rest being β-Si_3N_4. The SiC powder (Ultrafine grade, Ibiden Co. Ltd., Ogaki, Japan) used has a 0.27μm mean particle size. The samples were prepared using a mixture of silicon nitride, with 20 wt% SiC powder and 8wt% Y_2O_3 as an additive powder. To this mixture, alcohol was added and blended completely for 48h. The mixture was placed in an evaporator in order to extract the solvent and then in a vacuum in order to produce a dry powder mixture. The mixture was subsequently hot-pressed at 2073 K and 35MPa for 2h in nitrogen gas. The details of the material used are explained in Ref.13. The crack-healed zone has a quite high monotonic bending strength up to ~1573K.

2.2 Test specimen and pre-crack

The hot-pressed material was then cut into test specimens measuring 3mm×4mm×20mm. Figure 1 shows the shape and dimensions of the test specimen. A semi-elliptical surface crack of 100μm in surface length, which reduces three-point bending strength by half, was introduced at the center of the tension surface of the test specimens using a Vickers indenter at a load of 19.6 N. The ratio of depth (a) to half surface length (c) of the crack (aspect ratio) was $a/c \doteqdot 0.9$.

2.3 Crack-healing process and subsequent experiments

A hydraulically controlled testing machine, equipped with electric furnace, was used for the crack-healing under cyclic stress. These tests were carried out in air using a three-point loading system with a span of 16mm (see Fig.1). The loading waveform was sinusoidal at a frequency (f) of 5Hz with stress ratio ($R=\sigma_{min}/\sigma_{max}$) of 0.2. In the crack-healing processes, we first applied a cyclic bending stress and then increased furnace temperature at a rate of 10℃/min in order to avoid unexpected crack-healing without stress. The specimens were subjected to cyclic bending stress at each healing temperature for a given time. The specimens were furnace-cooled to room temperature. After the specimens had cooled completely, the cyclic stress was removed followed by monotonic bending tests or cyclic fatigue tests.

The loading system in monotonic bending tests and cyclic fatigue tests was also three-point bending with a span of 16mm. Monotonic bending tests were carried out at room temperature or at each healing temperature using universal testing machine, equipped with electric furnace. The cross-head speed was 0.5mm/min. Cyclic fatigue tests were conducted at each healing temperature with sinusoidal waveform at a stress ratio (R) of 0.2 and a frequency of 5Hz. The maximum value of applied stress at which a specimen did not fracture up to 10^6 cycles was defined as the cyclic fatigue limit, σ_{f0}.

Table 1 shows the conditions of the crack-healing and subsequent testing. Experiments (i) and (ii) were carried out in order to determine the threshold stress for crack-healing at 1273 and 1473 K. A pre-cracked specimen was crack-healed under cyclic stress at the temperatures of 1273 and 1473 K for 5h in air. After the crack-healing, monotonic bending tests were carried out at room temperature. Then, we defined the threshold stress for crack-healing.

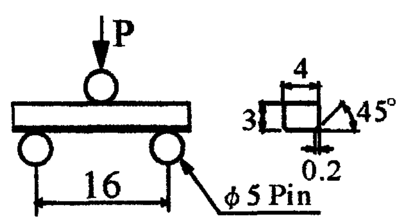

Fig.1 Shape of specimen and loading system; in mm.

If a material could heal a crack under service condition, i.e. under stress and at service temperature, and if the healed zone were strong enough at service temperature, it would be very beneficial. In order to investigate this possibility monotonic bending tests and cyclic fatigue tests of specimens crack-healed under cyclic stress were carried out at healing temperatures between 1173 and 1473 K (see experiments (iii) to (vi) in Table 1).

The fracture initiation site was identified by optical microscope. The fracture surfaces and specimen surfaces were analyzed using a scanning electron microscope (SEM).

3. RESULTS AND DISCUSSION

3.1 Threshold stress for crack-healing

Figure 2 shows the bending strength of crack-healed specimens as a function of applied stress during crack-healing. In Fig.2, room-temperature bending strengths of as-received smooth specimens (○) and pre-cracked specimens (△) are also indicated on the left-hand side. The average value of the bending strength of smooth specimens was ~800MPa. The pre-crack largely reduced the bending strength to ~400MPa.

Below applied stress σ_{ap}=300MPa, all specimens survived the cyclic stress for 5h. The bending strength recovered and most specimens fractured outside the crack-healed zone, indicating the pre-cracks were healed completely. There is only a small difference in bending strength over the range of applied stress up to 300MPa.

At σ_{ap}=350MPa, mixed behavior was observed, where four specimens survived the cyclic stress for 5h, while three others shown by symbols ▲ fractured up to ~1000 cycles (~200s) before reaching the healing temperature. Thus, we defined the threshold stress for crack-healing, below which a pre-cracked specimen recovered its bending strength, as 300MPa. It was surprising that a pre-crack of considerable length (2c=100μm) could be healed completely even under a quite high cyclic bending stress of 300MPa, which was a

Table 1 Conditions for crack-healing and subsequent experiments.

No.	Crack healing conditions			Experimental conditions	
	Temperature T_H (K)	Time t_H (h)	Applied stress σ_{ap} (MPa)	Temperature (K)	Testing type
i	1273	5	0, 210, 300, 350	Room temperature	Monotonic bending
ii	1473	5	0, 210, 300, 350	Room temperature	Monotonic bending
iii	1173	70	210	1173	Monotonic bending Cyclic fatigue
iv	1273	15, 50	210	1273	Monotonic bending Cyclic fatigue
v	1373	15	210	1373	Monotonic bending Cyclic fatigue
vi	1473	5	210	1473	Monotonic bending Cyclic fatigue

75% of the bending stress of the pre-cracked specimens (400MPa).

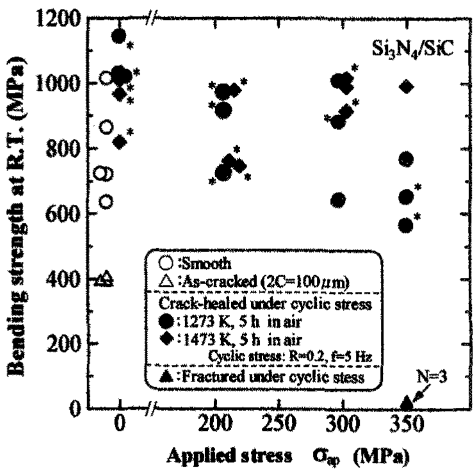

Fig.2 Relationship between applied stress during crack-healing and bending strength at room temperature. Data marked with an asterisk indicate that fracture occurred outside of the crack-healed zone.

3.2 Mechanism of crack-healing

The crack-healing of silicon nitride ceramics occurs due to oxidation. The estimated crack-healing reactions for Si_3N_4-SiC-Y_2O_3 are as follows [4,13];

$$Si_3N_4 + 3O_2 \rightarrow 3SiO_2 + 2N_2 \quad (1)$$

$$SiC + 2O_2 \rightarrow SiO_2 + CO_2 (CO) \quad (2)$$

$$2SiC + Y_2O_3 + 4O_2 \rightarrow Y_2Si_2O_7 + 2CO_2 (CO) \quad (3)$$

Formation of the SiO_2 and $Y_2Si_2O_7$ phases was confirmed by X-ray diffraction (XRD) analysis [12]. A crack is filled and bonded by SiO_2 and $Y_2Si_2O_7$.

Figure 3 shows a fracture surface of the specimen crack-healed at 1473 K for 5h under a cyclic stress of 350MPa. Although the specimen fractured from the crack-healed zone, the specimen recovered its bending strength. The initial crack front is denoted by the white solid line. The fracture surface of the crack-healed zone appears dark since the crack surface of the crack-healed zone oxidized. Therefore, it is clear that crack growth from the initial crack occurred under cyclic bending stress. The crack front that increased under the cyclic bending stress is denoted by white dotted line in Fig.3. However, crack-healing proceeded simultaneously; thus, the crack growth was retarded and eventually the crack was healed.

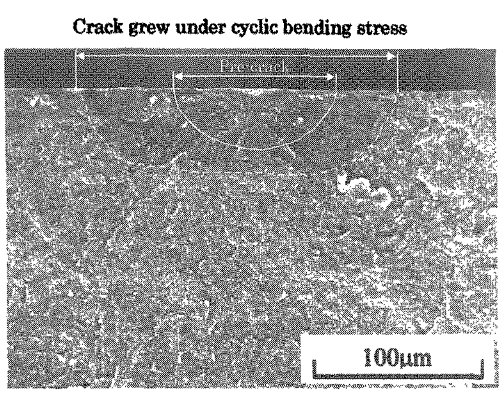

Fig.3 SEM micrograph of fracture surface of a test specimen subjected to a crack-healing under cyclic bending stress followed by a bending test at room temperature. σ_B=990MPa (Healing condition: 1473K, 5h in air, $\sigma_{max,ap}$=350MPa, R=0.2, f=5Hz).

3.3 Cyclic fatigue strength of crack-healed specimens

Figures 4(a) and (b) show the results of cyclic fatigue tests together with the monotonic bending strengths at crack-healing temperatures of 1173 and 1273 K, respectively. The monotonic bending test results are shown on the left-hand side of the figures. The symbol ■ in Fig. 4(a) shows the results of the specimens crack-healed at 1173K for 70h. The symbols ▲ and △

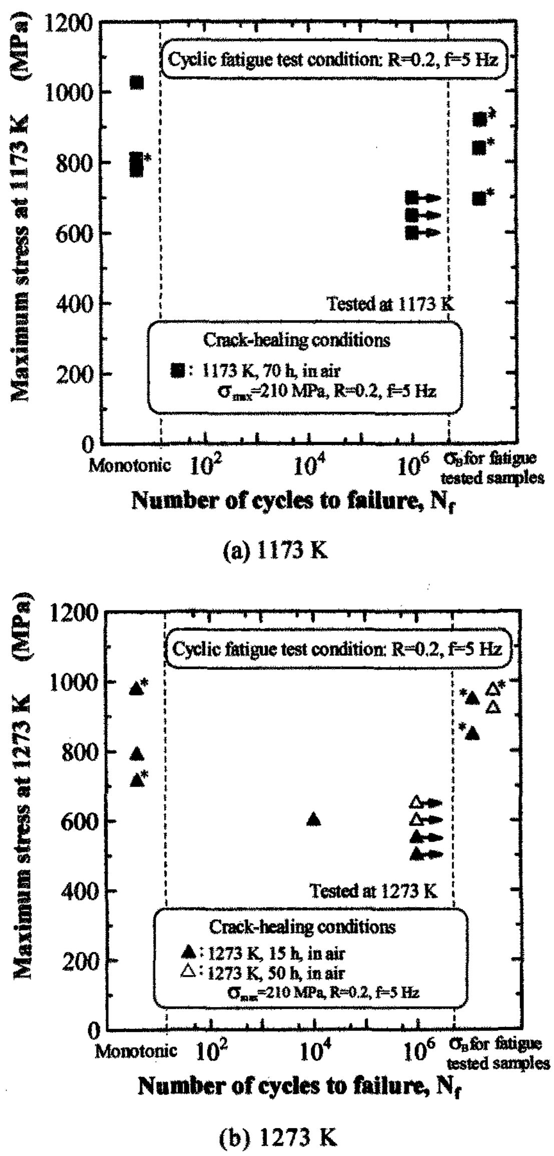

Fig.4 Results of bending tests and cyclic fatigue tests of Si_3N_4/SiC at healing temperatures between 1173 and 1273 K. Cyclic stress during crack-healing: σ_{max}=210MPa, R=0.2 and f=5Hz.

in Fig. 4(b) shows the results of the specimens crack-healed at 1273K for 15h and 50h, respectively. Asterisks show that the fracture occurred outside the crack-healed zone. The average values of monotonic bending strength ($\sigma_{B,ave}$) for 1173 K (■) and 1273 K crack-healed specimens (▲) are 872MPa and 826MPa, respectively, at each healing temperature. The bending strength of crack-healed specimens is comparable to the room-temperature bending strength of smooth specimens, i.e., 800MPa.

The specimens that did not fracture in the cyclic fatigue tests are marked by arrow symbols (→). The cyclic fatigue limit (σ_{f0}) for the 1173 K crack-healed specimen was 700MPa at 1173 K. The σ_{f0} for the specimens crack-healed at 1273 K for 15h was 550MPa, as shown by symbol ▲. Extending the healing time from 15h to 50h, the σ_{f0} increased up to 650MPa. The cyclic fatigue limit was considerably high. The bending strength of the specimens which survived the cyclic fatigue tests were also investigated and shown on the right-hand side of Figs. 4(a) and (b). The bending tests were conducted at each crack-healing temperature, i.e., 1173 and 1273 K. The fatigue-tested specimens exhibited bending strengths similar to the monotonically tested specimens, indicating that significant crack growth from the crack-healed zone or internal defects did not occur.

Figure 5 shows the bending strength and cyclic fatigue limit at healing temperatures between 1173 and 1473 K. The ratio of σ_{f0} to the average bending strength ($\sigma_{B,ave}$), $\sigma_{f0}/\sigma_{B,ave}$, was in the range between 0.67 and 0.86 at each healing temperature. Therefore, the cyclic fatigue limit was close to the bending strength. We presume that the creak-healing also occurred during crack-healing. Thus, the values of cyclic fatigue limit were quite high.

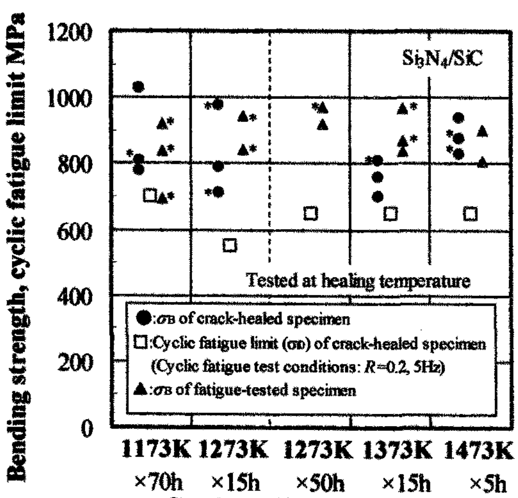

Fig.5 Results of bending tests and cyclic fatigue tests of Si_3N_4/SiC at healing temperatures between 1173 and 1473 K. Cyclic stress during crack-healing: σ_{max}=210MPa, R=0.2 and f=5Hz.

4. CONCLUSIONS

Si_3N_4/SiC composite ceramics were hot-pressed and subjected to three-point bending. A semi-elliptical surface crack of 100μm in surface length was made on each specimen. The pre-cracked specimens were crack-healed under a cyclic bending stress of 5 Hz, and the resultant bending strength and fatigue strength were studied. The threshold stress for crack-healing was investigated at 1273 K and 1473 K. The cyclic fatigue strength of crack-healed specimens was investigated at healing temperatures of 1173 and 1473 K.

(1) The threshold stress for crack-healing, below which a pre-cracked specimen recovered its bending strength, was 300MPa at healing temperatures of 1273 and 1473 K. The threshold stress of 300MPa was 75% of the bending stress of the pre-cracked specimens (~400MPa). Surprisingly, a pre-crack of considerable length ($2c$=100μm), which reduced the bending strength by half, could be healed completely even under quite high cyclic bending stress.

(2) The cyclic fatigue limits of the specimens crack-healed under cyclic stress at healing temperatures between 1173 and 1473 K are between 550MPa and 650MPa at each healing temperature. The ratio of σ_{f0} to the average bending strength ($\sigma_{B,ave}$), $\sigma_{f0}/\sigma_{B,ave}$, was in the range between 0.67 and 0.86 at each healing temperature. Thus, the cyclic fatigue limit of crack-healed specimens is considerably high.

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