Effects of Frequency on the Crack-Healing Behavior of \( \text{Si}_3\text{N}_4/\text{SiC} \) Composite under Cyclic Stress

Koji TAKAHASHI**, Yuuta MIZOBE***, Kotoji ANDO** and Shinji SAITO****

\( \text{Si}_3\text{N}_4/\text{SiC} \) ceramics were hot-pressed to investigate the effect of stress frequency on the crack-healing behavior under cyclic stress. The specimens having pre-crack of 100 \( \mu \)m were subjected to crack-healing under constant or cyclic bending stresses of 300 MPa at temperatures between 800 and 1200 \( \degree \)C. The resultant bending strength of the crack-healed specimen was investigated. At a healing temperature of 900 \( \degree \)C, the pre-cracks had been completely healed both under constant stress and cyclic stress of 0.5 Hz. However, the pre-cracks had not completely healed under a cyclic stress of 5 and 10 Hz. Thus, the stress frequency affects crack-healing behaviors at 900 \( \degree \)C. On the other hand, when the healing temperature was higher than 1000 \( \degree \)C, the pre-cracks were healed completely under both constant stress and cyclic stress up to 10 Hz. The effects of stress frequency were dependent on the healing temperature. The reasons for this were discussed from the viewpoint of competitive mechanism of fatigue crack growth and crack-healing.

Key Words: Ceramics, Crack Propagation, Crack-Healing, Silicon Nitride

1. Introduction

Silicon nitride based ceramics and composites are candidate materials for industrial applications because of their excellent mechanical, tribological, and thermal properties. These applications include turbo charger rotors, diesel engine components, cutting tools, bearings and coil springs. Engineering ceramics, including \( \text{Si}_3\text{N}_4 \), have a self-crack-healing ability(1)–(7). In engineering applications, this self-crack-healing ability may increase the reliability of ceramic structural members, thereby decreasing the inspection, machining, and polishing costs of maintaining these components.

We investigated the crack-healing behaviors of silicon nitride reinforced by silicon carbide (\( \text{Si}_3\text{N}_4/\text{SiC} \))(7)–(12). Excellent crack-healing ability can be induced by dispersing SiC particles to a matrix of \( \text{Si}_3\text{N}_4 \). Semi-elliptical surface cracks as long as 100 \( \mu \)m in surface length can be completely healed by simple heat-treatment in air. The optimal heat treatment condition for healing such cracks is 1300 \( \degree \)C for 1 h in air. These surface cracks were healed by an oxidation reaction when the crack surfaces were oxidized in reacting to the air. After heat-treatment in air, the bending strength of pre-cracked specimens recovered to a value similar to that of smooth specimens because the bonding force acting across the crack surfaces increased substantially. The crack-healed specimens showed excellent high-temperature strength and static or cyclic fatigue strengths(8), (9).

To increase the reliability of ceramic components, we proposed a methodology called the "crack-healing + proof test"(13). Generally, many surface cracks are induced by machining, decreasing its reliability considerably. However, surface cracks can be healed completely by crack-healing under optimized conditions. However, oxygen is necessary for such crack-healing. Consequently, an embedded crack cannot be healed. This fact indicates that the structural integrity of ceramic components cannot be guaranteed before service by crack-healing alone. Thus proof testing is necessary. Before service, the structural integrity of ceramic components can be increased by using the concept: crack-healing + proof test(13), (14).

Ceramic components are often operated under continuous loading with several frequencies, at elevated temperatures. If a crack initiated during service, the component’s
reliability are reduced considerably. If the crack could be healed under service conditions, and the healed zone has sufficient strength, the reliability and lifetime of ceramic components can be increased. From this view point, we have studied the crack-healing behaviors of Si$_3$N$_4$/SiC under constant and cyclic stress and found that pre-cracks of 100$\mu$m could be healed completely, even under constant or cyclic stress$^{(11)}$. It is well known that the frequency of cyclic stress significantly affects the fatigue strength of Si$_3$N$_4$ base material at room temperature$^{(15)}$–$^{(17)}$. The Si$_3$N$_4$ base material can be toughened by elongating the grain structure to promote intergranular fracture. However, under cyclic stress, toughening mechanisms such as crack shielding and crack bridging are degraded by frictional wear along the crack surfaces. Therefore, as the frequency of cyclic stress increases, the fatigue crack growth rate accelerates, leading to decreased cyclic fatigue strength. The crack-healing behavior under stress is determined by the competitive mechanism between fatigue crack growth and crack-healing$^{(12)}$. Therefore, it is important to investigate the effects of frequency on the crack-healing behavior of the Si$_3$N$_4$/SiC. However, the effect of frequency on the crack-healing behavior of Si$_3$N$_4$/SiC has not been studied before.

From this perspective, the effects of stress frequency on crack-healing behavior of Si$_3$N$_4$/SiC were investigated in this study. The specimens having pre-cracks of 100$\mu$m were subjected to crack-healing under constant or cyclic stress at temperatures between 800 and 1 200$^\circ$C. The resultant bending strength of the crack-healed specimen was investigated. The effects of stress frequency were discussed from the viewpoint of the competitive mechanism between fatigue crack growth and crack healing behavior.

2. Experimental Procedures

2.1 Material, test specimen and pre-cracks

The silicon nitride powder (SN-E10, Ube Industries Ltd., Ube, Japan) used in this study has a mean particle size of 0.2$\mu$m; the volume ratio of $\alpha$-Si$_3$N$_4$ is about 95%, the rest being $\beta$-Si$_3$N$_4$. The SiC powder (Ultrafine grade, Ibiden Co., Ltd., Ogaki, Japan) used has a 0.27$\mu$m mean particle size. The samples were prepared using a mixture of silicon nitride, with 20 wt.% SiC powder and 8 wt.% Y$_2$O$_3$ as an additive powder. The Y$_2$O$_3$ powder (Fine grade, Nippon Yttrium Co., Ltd., Oomuta, Japan) used has a 0.4$\mu$m mean particle size. To this mixture, alcohol was added and blended completely for 48 h. The mixture was placed in an evaporator to extract the solvent and then in a vacuum to produce a dry powder mixture. The mixture was subsequently hot-pressed at 1 800$^\circ$C and 35 MPa for 2 h in nitrogen gas. The relative density of the hot-pressed material determined by the Archimedes method was 99.5%. Most SiC particles are located in grain boundaries and distributed uniformly.

The hot-pressed material was cut into test specimens measuring 3 mm $\times$ 4 mm $\times$ 20 mm. These specimens were subjected to three-point bending with a span of 16 mm. Semi-elliptical surface cracks of 100$\mu$m in surface length were introduced at the center of the tensile surface of the 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 $a/c \approx 0.9$.

2.2 Crack-healing process and bending tests

In this study, crack-healing was carried out under constant or cyclic bending stress using a hydraulically controlled testing machine equipped with an electric furnace. Figure 1 schematically shows the process of crack-healing adopted in this study. The waveform of cyclic bending stress was sinusoidal at a frequency of 0.5, 5 and 10 Hz with a stress ratio ($R = \sigma_{\text{min}}/\sigma_{\text{max}}$) of 0.2. The value of constant stress ($\sigma_{\text{ap}}$) and maximum stress ($\sigma_{\text{max,ap}}$) was 300 MPa. In order to avoid unexpected crack-healing without stress, we first applied a cyclic bending stress and then increased the furnace temperature at a rate of 10$^\circ$C/min and maintained the healing temperature at 800, 900, 1 000 and 1 200$^\circ$C for 5 h in air. The specimens were furnace-cooled to room temperature. After the specimens had cooled completely, the bending stress was removed.

After crack-healing process, monotonic bending tests were carried out at room temperature. For a comparison, bending tests for smooth specimens (heat-treated in air at 1 300$^\circ$C for 1 h) and pre-cracked specimens were also carried out. Normally, three specimens were used to establish the monotonic bending strength. These tests were carried out using a universal monotonic testing machine. The cross-head speed for the monotonic bending tests was 0.5 mm/min.

2.3 Direct observation of crack-healing process

In this study, direct observations of crack-healing process were carried out at 800, 900, 1 000 and 1 200$^\circ$C for 1 h in air to investigate the effects of healing temperature on crack-healing process. High-temperature micro-
scope system which consists of heating stage (maximum operating temperature 1400°C) and optical microscope (maximum magnification × 2500 in video monitor) were used. The motion pictures of crack-healing behavior were recorded to hard disk of personal computer.

2.4 Fatigue tests of pre-cracked specimens

The static and cyclic fatigue tests of the pre-cracked specimens were carried out in air at room temperature in order to examine the fatigue crack growth behavior of the pre-cracked specimens. These tests were carried out using a hydraulically controlled testing machine. This testing machine is identical to that used in the crack-healing under stress. The wave form of cyclic stress was sinusoidal at a stress ratio \((R)\) of 0.2 and a frequency \((f)\) of 0.5, 5 and 10 Hz. The static fatigue limit \((\sigma_{f0})\) and cyclic fatigue limit \((\sigma_{f0})\) were defined as the maximum stress under which specimens endured \(5 \times 10^5\) seconds (139 h).

3. Experimental Results

3.1 Effects of stress frequency on crack-healing behavior at 800 to 1200°C

The pre-cracked specimens were subjected to crack-healing under constant or cyclic bending stresses of 300 MPa in air at temperatures between 800 and 1200°C. Then, the bending strength of the crack-healed specimen was investigated in air at room temperature. Figure 2 shows the resultant bending strength as a function of crack-healing temperature. In Fig. 2, the bending strengths at room temperature for smooth specimens heat-treated at 1300°C for 1 h (open circle) and pre-cracked samples (open triangle) are also indicated. The mean value of the bending strength of heat-treated smooth specimens is 890 MPa. The Vickers indentation largely reduced bending strength to 400 MPa. At the crack-healing temperature of 800°C, the bending strength of the crack-healed specimens didn’t recover indicating that the pre-crack had not been completely healed for any frequency tested. This is because the oxidation reaction of Si₃N₄/SiC which contributes to crack-healing was not promoted at 800°C as will be mentioned in the next section. At the crack-healing temperature of 900°C, the bending strength of specimens crack-healed under constant stress recovered completely. However, bending strength of the specimens crack-healed under cyclic stress of 5 Hz and 10 Hz did not recover. At healing temperatures of 1000 and 1200°C, the specimens recovered their bending strength. The asterisk symbol shows that a fracture occurred outside the crack-healed zone indicating that the pre-crack had healed completely. Many specimens crack-healed at 1000 and 1200°C fractured outside the crack-healed zone, indicating the pre-crack was healed completely.

In order to investigate the effects of stress frequency at 900°C, the pre-cracked specimens were subjected to crack-healing under stress frequency of 0.5 Hz. Figure 3 shows the effects of stress frequency on the bending strength for specimens crack-healed under constant stress (open diamond) or cyclic stress (solid circles). The bending strengths of specimens crack-healed without applied stress (solid square) are also indicated. As already mentioned, the bending strength of those specimens crack-healed under cyclic stress of 5 and 10 Hz was not completely recovered. However, the bending strengths of specimens crack-healed under a cyclic stress of 0.5 Hz showed similar bending strength to those crack-healed under constant stress or no stress. Thus, it can be said that the pre-crack can be healed if the frequency of cyclic stress is less than 0.5 Hz. The stress frequency affected the crack-healing behavior at 900°C. This reason will be discussed in section 3.4 based on the experimental results mentioned.
3.2 Direct observation of crack-healing process

The pre-cracked zone was observed directly with optical microscope during heat-treatment process at 800, 900, 1000 and 1200°C in air to investigate of effects of healing temperature on crack-healing process.

Figure 4 shows the crack-healing process at 1000°C. Before crack-healing, pre-cracks can be clearly observed, as shown in Fig. 4 (a). The pre-cracks can not be clearly distinguished 30 min after it reached 1000°C, as shown in Fig. 4 (b). The pre-cracks could barely observed after 1 h at 1000°C as shown in Fig. 4 (c) because the surface was covered with newly created oxide (SiO₂ or Y₂Si₂O₇)(11),(12). At healing temperature of 1 200°C, oxidation reaction proceeded more quickly than the case of 1000°C. Figure 5 shows the crack-healing process at 900°C. Before crack-healing, pre-cracks can be clearly observed, as shown in Fig. 5 (a). The pre-cracks were gradually getting unclear with time. Thus, crack-healing occurred at 900°C. However, oxidation reaction proceeded more slowly than the case of 1000°C. Based on above mentioned results, it is apparent that the crack-healing are promoted at temperature above 1000°C.

3.3 Fatigue strength of the pre-cracked specimen

Static or cyclic fatigue tests were carried out to investigate the fatigue crack growth behavior of the pre-cracked specimens. Figure 6 (a) shows the results of the fatigue tests. The ordinate axis indicates applied stress (σ_{ap}) for static fatigue tests or maximum stress (σ_{max}) for cyclic fatigue tests. The abscissa axis indicates time to failure (t_f). The specimens that did not fracture in the static fatigue tests up to 5×10⁷ s are marked by arrow symbols (→). The static fatigue limit (σ_{f0}) shown by the open diamond was determined to be 325 MPa. The cyclic fatigue limit (σ_{f0}) for pre-cracked specimens tested at 0.5 Hz (solid reverse triangle) and 5 Hz (solid circle) was 275 MPa. The σ_{f0} for
the pre-cracked specimens tested at 10 Hz (solid triangle) was 250 MPa. As the frequency of the cyclic stress increased, fatigue limits ($\sigma_{f0}$) at $5 \times 10^5$ s and time to failure ($t_f$) at a given stress decreased. Thus, the Si$_3$N$_4$/SiC composite showed cyclic fatigue degradation. Cyclic fatigue degradation in silicon nitride was reported by several researchers$^{(15)}$–$^{(17)}$. Toughening mechanisms such as crack shielding and crack bridging can be degraded by cyclic stress. Figure 6 (b) shows the arrangement of the results of cyclic fatigue tests. The relationship between maximum stress ($\sigma_{max}$) and number of cycles to failure ($N_f$) is indicated. The relationship between $\sigma_{max}$ and $N_f$ is almost coincident for several frequencies. Thus, it is apparent that the fatigue behavior of Si$_3$N$_4$/SiC at room temperature is cyclic dependent rather than time-dependent.

3.4 Mechanism of crack growth and crack-healing

The mechanisms of crack-healing under constant or cyclic stress are discussed from the viewpoint of crack growth and crack-healing. Firstly, the crack-healing behavior at 900°C is discussed. In the case of crack-healing under constant stress, the value of the applied stress (300 MPa) is lower than the static fatigue limit (325 MPa) of the pre-cracked specimens. Thus, it is assumed that the pre-cracks are easily healed because crack growth from a pre-crack does not occur.

In the case of crack-healing under a cyclic stress of 0.5 Hz, the value of the applied stress (300 MPa) is slightly higher than the cyclic fatigue limit (275 MPa) of the pre-cracked specimens. However, the crack growth rate under 300 MPa is small because the time to failure ($t_f$) of the pre-cracked specimen is quite long if the frequency is 0.5 Hz as shown in Fig. 6 (a). Thus, it is assumed that the pre-cracks are healed because crack growth from a pre-crack is small.

In the case of crack-healing under a cyclic bending stress of 5 Hz or 10 Hz, the value of the applied stress (300 MPa) is higher than that of the cyclic fatigue limit (250 or 275 MPa) of the pre-cracked specimens. Thus, crack growth from the pre-crack tip occurred as will be described below. Figure 7 shows SEM micrographs of the fracture surface of the specimen crack-healed at 900°C under a cyclic bending stress of 5 Hz followed by a bending test at room temperature. The pre-crack front made by Vickers indentation is denoted by the white solid line in Fig. 7. The crack front that propagated under the cyclic bending stress is denoted by a white dotted line. The crack-healed zone appears dark because the crack-healed zone is oxidized$^{(12)}$. It is clear that crack growth from pre-cracks occurred extensively during the crack-healing process. Oxidation reaction is not promoted at 900°C, as mentioned in section 3.2. Thus, the cracks propagated under cyclic stress of 5 and 10 Hz were not completely healed at 900°C.

At the healing temperatures of 1 000 and 1 200°C, the pre-cracks are easily healed under constant stress. In the case of crack-healing under a cyclic stress of 5 and 10 Hz, it is assumed that crack growth from the pre-crack occurs. However, at higher temperatures such as 1 000 and 1 200°C, oxidation reactions are promoted. Therefore, it is assumed that the crack growth rate decreased due to crack-healing, and the cracks were eventually healed.

4. Conclusions

Si$_3$N$_4$/SiC composite ceramics having excellent crack-healing ability were hot-pressed in order to investigate the effect of stress frequency on crack-healing behavior under stress. The pre-cracked specimens were subjected to crack-healing under constant or cyclic bending
Fig. 7  SEM micrographs of fracture surface of Si₃N₄/SiC subjected to crack-healing under cyclic stress followed by bending test at room temperature. Crack growth occurred during crack-healing (Healing conditions: 900°C, 5 h in air, σ_{max,ap} = 300 MPa, R = 0.2, f = 5 Hz, bending strength at R.T. = 557 MPa)

stresses of 300 MPa in air at temperatures between 800 and 1200°C. The resultant bending strength of the crack-healed specimen was investigated. The fatigue tests of the pre-cracked specimens were also carried out in order to investigate fatigue crack growth behavior. Based on our test results, we obtained the following conclusions:

1) The pre-cracked specimens of Si₃N₄/SiC composite showed high sensitivity to cyclic stress. As the frequency of the cyclic stress increased, fatigue limits determined at 5 × 10⁷ s and time to failure at given stress decreased.

2) Direct observation of crack-healing process indicated that the oxidation reaction which contributed to the crack-healing of Si₃N₄/SiC composite were promoted at healing temperatures above 1000°C.

3) The stress frequency affected the crack-healing behaviors at 900°C. The pre-cracks were completely healed both under constant stress and cyclic stress of 0.5 Hz. However, the pre-cracks were not completely healed under a cyclic stress of 5 and 10 Hz. Fractographic observation indicated that the crack extension from pre-cracks occurred extensively during the healing process when the stress frequency was higher than 5 Hz. Thus, the extended cracks were not completely healed at 900°C.

4) The pre-cracks were healed completely under cyclic stress up to 10 Hz at healing temperatures of 1000 and 1200°C. Thus, the stress frequency didn’t affect crack-healing behaviors at 1000 and 1200°C. The cracks extended under cyclic stress at 5 and 10 Hz could be completely healed because the crack-healing of Si₃N₄/SiC were promoted at healing temperatures above 1000°C.

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