CRACK HEALING BEHAVIOUR OF Si_3N_4 CERAMICS AND ITS APPLICATION TO STRUCTURAL INTEGRITY

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The crack healing behaviour of two kinds of Si_3N_4/SiC composites with different sintering additives, and monolithic Si_3N_4 is studied. Strength of specimens was reduced to 43-48 % of that of smooth specimens by semi-circular surface crack. The crack size was about 100 μm in diameter. However, the strength recovered completely by healing. The best healing condition depends on chemical composition. For sample SN-S27-Y8, the best healing condition was found to be 1200℃, for 1h and in air. The crack healed specimen at the best condition showed sufficient strength up to 1400℃. The benefit of [proof test +healing] was studied by using coil springs made of silicon nitride, systematically. It was verified that [proof test + healing] was an excellent technique for the structural integrity of ceramics up to high temperatures.

INTRODUCTION

Structural integrity is a very important concept for the safety and high performance of ceramics. Fracture toughness of ceramics is not so high and ceramics are sensitive to flaws. It is very difficult to keep a high level of integrity, because their allowable flaw size is very small. To overcome this difficulty, a proof test is developed. However, it is not enough for the complete structural integrity. Recently, it was found that some ceramics have crack healing ability(1-3). If the both techniques are applicable to ceramics, it will be possible to overcome the difficulty. However, the most important issues as to crack healing were not studied. The issues are: (a)effect of chemical compositions on healing ability, (b)effect of healing condition on strength, (c)crack healed sample has sufficient strength properties or not, (d)proposed [proof test + healing]is an excellent technique even at high temperature or not. In this paper, above three issues[b,c,d] were studied by using three kinds of silicon nitride ceramics and ceramic coil springs. It was concluded that proposed [proof test +healing] is an excellent technique for structural integrity of ceramics.

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SPECIMEN AND EXPERIMENTAL PROCEDURE

Material

Bend specimen Silicon nitride powder(Grade SN-E10, UBE Industries, Japan) and SiC power(Grade Ultrafine, Ibiden Industries, Japan, mean particle size:0.27 μm) were used in this investigation. Sintering additives were added Si_3N_4 powder and then the powders were mixed in alcohol for 48 h using a nylon ball mill. After drying them, the powders were hot pressed(HP). The hot pressing was carried out in 35MPa N_2 gas at 1850°C. In this study, three kinds of specimens were prepared, see TABLE 2. Test specimens (3 x 4 x 40mm) were cut and machined from the sintered samples(90 x 90 x 5 mm) and surface of the test specimen were ground and polished before testing in accordance with the Japan Industrial Standard(JIS)(4).

Coil spring. The Si_3N_4 powder(SN-E10) was used in this work. 5wt% Y_2O_3 and 3wt% Al_2O_3 were used as a sintering additives. These powder were mixed and then dried. As binders, methyl cellulose was used. The mixture was formed into a wire via an extruder. The wire material obtained was coiled into springs, and the burning off of organic binders and sintering was then performed. Sintering was carried out in 0.93 MPa N_2 gas for 4 h at 1850°C. Finally, the ends of the springs were shaped by using diamond wheel grinder. And no other mechanical grinding was made on coil spring specimen. The size of coil spring are given in Table.1

TABLE 1 — Dimension of Si_3N_4 Coil Spring.

Wire Diameter [mm]	Mean Coil Diameter [mm]	Number of Active Coils
ϕ 2.4	ϕ 20.65	13

Experimental Method

A semi-circular crack was introduced at the center of the tensile surface of the bend specimen using Vickers hardness tester. Indentation load and crack diameter were 19.6 N and about 100 μm, respectively.

To investigate the effect of healing condition on the crack healing ability, a healing temperature of 800°C - 1400°C and a healing time of 1h-150h in air were used.

To evaluate bending strength of smooth or cracked specimen, four-point bending test was conducted(outer support span: 30mm, inner load span:10mm, cross head speed: 0.5mm/min) following the JIS1601 method(4). Bending strength was measured from at room temperature to 1400°C in air. Five specimens were used for room temperature test.

However, three specimens were used for the test at high temperature, usually.

Compression test (cross head speed : 20mm/min) was carried out to evaluate shear fracture stress of Si_3N_4 coil spring. Proof test was carried out by stressing up to 417 MPa at room temperature and the cross head speed of the test was 20 mm/min.

To evaluate static fatigue properties of crack-healed specimen, slow strain rate test was carried out. The specimen were indented and subsequently they were crack-healed at 1300℃ for 1 h in air. Then specimen were tested at three level cross head speed of 0.5, 0.01 and 0.001mm/min by using three point bend test system. Test temperature was 1300℃ and 1400℃.

Scanning electron microscope was used to observe the morphology of crack and surface of crack-healed specimens. X-ray diffraction analysis was used to identify the surface oxides phase present on the specimen which was crack-healed at different conditions. Laser microscope was used to observe crack healing process, directly.

TEST RESULTS AND DISCUSSION

Effect of Healing Condition on Strength at Room Temperature

Table 2 shows the effect of healing conditions(temperature and time) on bending strength and X-ray diffraction results of surface oxides of crack-healed specimen. The strength of as-cracked specimen was reduced to 43-48% of that of smooth specimen. However, the strength recovered by crack healing treatment in air. The recovery ratio depends on healing condition and sample. In case of SN-Y5A3, when specimens healed a crack at 1100℃ or 1200℃, their strength were recovered completely. Specimen crack-healed at 1000℃ and 1300℃ showed a little lower strength than that of smooth specimen. It can be concluded, from these results, that the optimum healing temperature for SN-Y5A3 is 1100℃ or 1200℃.

As the healing temperature in air increased, in case of SN-S27-Y5A3, the strength increased gradually. By the healing at 1200℃, the strength of cracked specimen recovered completely and showed a maximum value of 868 MPa. However, by the healing at 1300℃, their strength was decreased slightly. In case of SN-S27-Y8, when specimens healed a crack at the temperature ranging from 900℃ to 1300℃, they recovered their strength completely. And most specimens failed at a location different from the healed crack. This means that crack-healed area have sufficient strength. By the healing at 1400℃, the strength become slightly lower than that of smooth specimen. From above facts, it can be seen that best healing temperature range for SN-S27-Y8 is wider than that of SN-Y5A3 and SN-S27-Y5A3. The increase in strength by crack healing in air at high temperature can be attributed to crack healing by oxidation. By SEM observation, it can be found that new materials were produced on surface of the crack-healed specimen. The crystalline phases of new products are listed in Table 2.

TABLE 2 — Results of Bending Test and X-ray Diffraction Analysis.

Sample Name	Sintering Additives	Crack Healing Condition [℃ × hr.]	Bending Strength at R.T [MPa] Smooth Specimen	As-Cracked Specimen	Crack Healed Specimen	Surface Oxides Phase
SN-Y5A3	$Y_2O_3$5wt% +$Al_2O_3$3wt%	1000 × 1	971	469	792	—
		1100 × 1			1059	
		1200 × 1			1064	$Y_2Si_2O_7$ SiO_2, Y_2SiO_5
		1300 × 1			897	
SN-S27-Y8	SiC20wt% +$Y_2O_3$8wt%	800 × 5	751	363	555	—
		900 × 5			745	
		1000 × 1			758	$Y_2Si_2O_7$ SiO_2
		1200 × 5			829	
		1300 × 1			792	
		1400 × 1			721	
SN-S27-Y5A3	SiC20wt% +$Y_2O_3$5wt% +$Al_2O_3$3wt%	800 × 150	856	371	586	—
		1000 × 1			600	
		1100 × 1			672	$Y_2Si_2O_7$ SiO_2
		1200 × 1			868	
		1300 × 1			753	

SN:Si_3N_4, S27:SiC(mean particle size 0.27 μm), Y:Y_2O_3, A:Al_2O_3

High Temperature Strength

Figure 1 shows that the effect of test temperature on bending strength of crack healed specimen. In case of SN-Y5A3(●), as the test temperature increased, the bending strength decreased gradually up to 1200 ℃. The strength of SN-SC27-Y5A3(□) shows a same value up to 1000℃. Above 1000℃, however, the strength decreased considerably with increasing test temperature. On the other hand, in case of SN-SC27-Y8(▲), the strength decreased slightly at 1100℃ and then did not exhibit any noticeable decrease in strength up to 1400℃.

Relationship between cross head speed and bending strength of SN-S27-Y8 are shown in figure 2. Test has been carried at 1300℃ and 1400℃. At both temperatures, the bending strength was not affected by cross head speed within the range tested. This facts indicate that crack healed area is not sensitive to static fatigue fracture.

Application

By above studies, it is shown that crack healing technique is a valuable method to assure structural integrity of member. In another paper, we proposed the effectiveness of proof test(5). If both techniques are superposed, the reliability of structural member will increase considerably. To investigate the possibility, fracture test has been made by using ceramic coil springs. Figure 3 shows the effects of proof test and healing treatment on the fracture stress of the Si_3N_4 coil spring. Also in figure 4, test temperature dependence of fracture stress of Si_3N_4 coil spring was shown. In both figures, "n" indicates the number of specimens which did not fracture by the compressive test and "N" indicates the total number of test specimens. For these test, proof stress(σ_p) was decided as 417MPa. Many tests have been made to decide the best healing condition of Si_3N_4 coil spring. In the result, it was found that the best healing conditions are 1100°C~1200°C, for 1hr. in air. From both figures, following characteristics were obtained:(a) The fracture stress of as-received coil spring showed wide scatter both at room temperature and 1100°C. (b) Generally, healed coils showed higher shear fracture stress than that of as-received coil. However, a few healed specimens showed very low fracture stress. This fact maybe caused by the fact that embedded flaws can not be healed. (c) In figure 4, increment of average shear fracture stress by treatment of [proof test + healing] is not significant for the temperature range tested. This is attributed to the fact that maximum loaded stress is adopted as a fracture stress of no-fractured specimen. (d) The lower fracture stress of [proof test + healing] specimen is remarkably higher than that of other series tests, especially at high temperature area. And the fracture stress is higher than the proof stress(417 MPa) , except for only one specimen. From these results, it can be concluded that the proposed combined method [proof test + healing] is a useful technique in improving the reliability of Si_3N_4 coil springs at R.T as well as at high temperature.

CONCLUSION

By heat treatment in air at high temperature, for example at 1200°C, the bend specimen with a semi-circular surface crack recovered their strength completely. This recovery of strength was attributed to crack healing. The new method [proof test + healing] is proposed and its usefulness for the structural integrity was verified by using coil spring, successfully.

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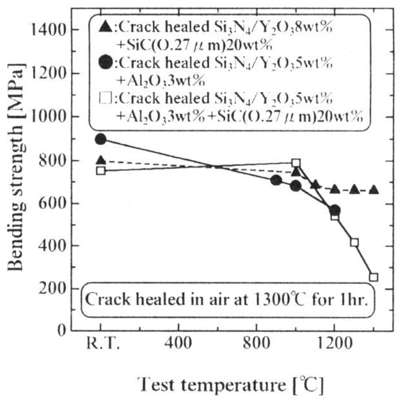

Figure 1 Effect of temperature on the bending strength of crack healed specimen.

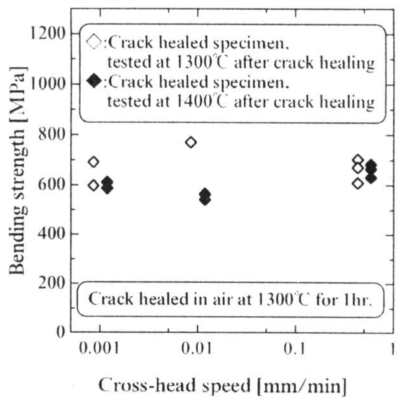

Figure 2 Relationship between cross-head speed and bending strength of SN-S27-Y8 ceramics with surface crack.

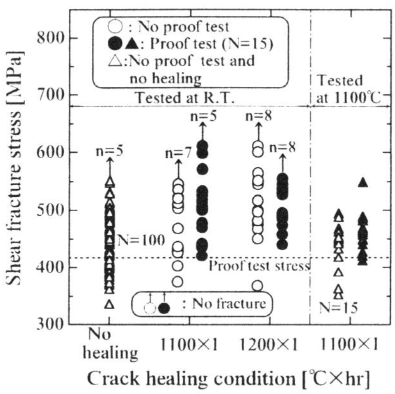

Figure 3 Relationship between crack healing condition and shear fracture stress of Si_3N_4 coil spring.

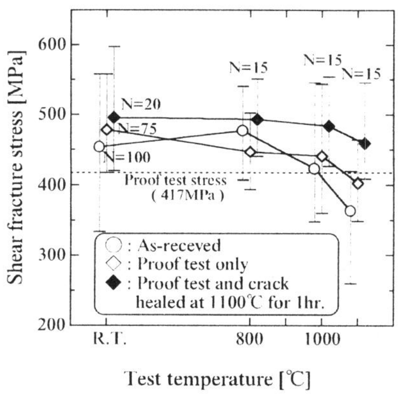

Figure 4 Temperature dependence of shear fracture stress of Si_3N_4 coil spring.