Fracture Energy and Tensile Strength of Silicon Nitride at High Temperatures

Tatsuki OHJI, Seisuke SAKAI, Masaru ITO, Yukihiko YAMAUCHI, Wataru KANEMATSU and Shoji ITO
(Government Industrial Research Institute, Nagoya, 1-1, Hirate-cho, Kita-ku, Nagoya-shi 462)

The work-of-fracture (WOF) tests were conducted for hot-pressed silicon nitride at high temperatures above 1000°C to investigate displacement rate (D-rate) dependence of effective fracture energy, and then the bridging stress at a crack interface was estimated from the increment of fracture resistance with crack extension (R-curve behavior) to discuss its correspondence to tensile strength. The effective fracture energy and the bridging stress increased with lowering D-rate, and then decreased, probably due to the activated pulling out work of grains and its sensitivity to change in D-rate. The bridging stress was shown to correspond to a great part of tensile strength in the range where a plasticity was seen in the stress-displacement (S-D) curve, implying a large contribution of grain bridging toward bearing an applied tensile stress during slow crack extension.

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1. Introduction

Silicon nitride, which is considered one of the most promising structural ceramics, exhibits non-elastic behavior in high temperature mechanical strength tests above 1000°C due to the softening of glassy phase formed at grain boundaries as a result of processing with sintering additives like alumina, yttria, magnesia, etc. [1]-[3] In accordance with lowering the viscosity of glassy phases at high temperatures, the cavities formed at grain boundaries or their triple points enhance subcritical crack growth, SCG, resulting in non-elastic stress-displacement, S-D, curves.

The toughening of non-transforming ceramics can be often attained by microcracking in the frontal process zone and grain bridging in the following wake region; the former sometimes accompanies the residual compressive stress in the wake region, leading to a rising R-curve behavior, the increase of fracture resistance with crack extension, as well as the latter case. [4]-[11] However, the softening of grain boundary glassy phase of silicon nitride at high temperatures is assumed to bring the similar effect of plastic deformation of dislocation, which will make it difficult to produce microcracks. [12] Hence, the increase in fracture toughness of silicon nitride at high temperatures, which has been reported by several researchers, [1],[13]-[16] is attributed to the grain bridging; the softening of glassy phase will give the appropriate bonding force between grains for the most activated pulling out works. Particularly the elongated silicon nitride grains are supposed to make a large contribution for it. This toughening mechanism is anticipated to bring about the rising R-curve behavior.

The high temperature fractures of silicon nitride are dependent on viscosity of glassy phase as well as grain morphology, etc, and, therefore, strongly affected by strain rate, or D-rate. Accordingly, the properties concerning fracture energy also should be dependent on D-rate. It was reported that in alumina containing glassy phase the effective fracture energy or the crack propag-
tion resistance heavily depended on D-rate at high temperatures, and the stress intensity factor took the maximum value at a certain D-rate.\textsuperscript{17,18} It was also known that fracture toughness of silicon nitride at high temperatures showed strong dependence on D-rate.\textsuperscript{19,20}

In this study, chevron notch bend, CNB, specimen, which facilitate stable fracture in brittle materials due to its triangular ligament configuration, is used to measure the work-of-fracture, WOF, of hot-pressed silicon nitride at high temperatures at various D-rate from 0.1 to 0.0001 mm/min, and the dependences of effective fracture energy and rising R-curve behavior on D-rate were discussed. Then, the bridging stress at the crack interface, which is estimated from the increment of fracture resistance against crack extension, is compared with tensile strength measured at the same temperature in air atmosphere.

2. Experimental procedures

Commercially available hot-pressed silicon nitride doped with 5 wt% yttria and 3 wt% alumina containing impurities of 0.007% Mg, 0.008% Cr, 0.032% Fe and 0.026% Ca (wt%) was used in this study. The density was determined to be 3.20 Mg/m\textsuperscript{3} by water immersion. Although some types of hot-pressed silicon nitride showed improvements of high temperature strength due to crystallization of grain boundary glassy phase into mellilite phase, etc.,\textsuperscript{21,22} Powder X-ray analysis of this material revealed little reflection from the mellilite one.

The dimensions and geometry of the CNB test specimen is shown in Fig. 1. The width of chevron notch was 100 μm, and three point bending tests with lower span of 30 mm were carried out. The most important problem in WOF tests is to use a stiff testing machine.\textsuperscript{23} As a matter of fact, in high temperature measurements, if loading rods, etc. are connected by a screw, the slack due to thermal expansion brings about degradation in stiffness of loading system. Hence in this experiment, a silicon carbide loading rod consisting of one body, which was linked with water cooling stainless rod outside the furnace, was used, and the slack at that connection by thermal expansion is eliminated by strictly tightening screws after the furnace was heated up to the test temperature. The other testing equipments inside the furnace were all made of silicon carbide. The “blank” tests without specimens were carried out to know the compliance of testing system for each temperature and D-rate, which corrected the load-deflection curves obtained in WOF tests into the true ones.

The tensile test specimen, whose geometry and dimensions are shown in Fig. 2, were fabricated from the same billets of hot-pressed silicon nitride. This type of specimen, where the elongation of gauge length between two arms was measured by optical extensometer (Zimmer OHG, Model 4100) through the silica glass windows mounted at both sides of furnace, was often used by several researchers so far,\textsuperscript{24,25} and the method of elongation measurements here was similar to them. The detailed tensile testing method was previously described.\textsuperscript{26}

Both tests were carried out in air atmosphere.
The furnace was heated to the test temperature in about one hour, and the specimen was held at this temperature for 30 min before the tests were started to achieve thermal equilibrium.

3. Results and discussion

The softening of grain boundary glassy phase of silicon nitride at high temperatures is known to result in the rapid decrease of mechanical strength. The temperature dependences of tensile strength and fracture toughness of this material, which were measured at $D$-rates of 0.1 and 0.01 mm/min, respectively, are shown in Fig. 3. The fracture toughness was determined from the maximum loads in CNB test as will be shown later. Tensile strength was degraded at a little higher temperature than 1000°C, where internal friction was rapidly raised with a peak, and elasticity began to drop.\(^{27}\)

3.1 Real and calculated crack lengths in CNB tests

Crack lengths during CNB tests can be calculated from the change in compliance using Bluhm’s slice model.\(^{29,30}\) This calculation requires as a premise that the material is an elastic body, and therefore its application into the material showing some plastic reaction sometimes leads to a gap between actual and estimated crack lengths. Bornhauser et al., succeeded in a direct measurement of crack length during notched beam bend tests at 1000°C using pure alumina and alumina containing glassy phase, and found the difference between measured and calculated crack lengths in the latter alumina.\(^{31}\) Thus, it is required to verify the applicability of the calculation methods into fractures of silicon nitride at high temperatures.

For this purpose, following experiments were conducted. At a certain halfway point of a WOF test at 1260°C, $D$-rate was changed from 0.01 to 10.0 mm/min to abruptly bring about an unstable fracture, as indicated by solid line of Fig. 4.
microscopic observation for the ligament portion revealed a border between stable crack growth wake at 0.01 mm/min and catastrophic one at 10.0 mm/min. Figure 5 shows a fracture surface of the example of Fig. 4. These operations were conducted at the various halfway points by use of several specimens, and as a result a real crack length curve against displacement was obtained. In Fig. 4, the real crack length curve against displacement measured at 1260°C and 0.01 mm/min is compared with the calculated one using Bluhm's slice model. There is a fairly good agreement between them. This method is applicable only when the load-displacement (L-D) curves of WOF test is completely reproducible, and for this reason, the identical notch introduction works into the specimens are strictly required.

3.2 Work-of-fractures

At 1260°C, WOF is not physical properties but are heavily dependent on D-rate. Figure 6 shows L-D curves at D-rates of 0.1, 0.01, 0.001 and 0.0001 mm/min at 1260°C. Figure 7 illustrates the dependences of effective fracture energy, γeff, and fracture toughness, KIC, on D-rate at 1260°C.

The γeff and KIC of Fig. 7 are defined as follows:

\[ \gamma_{eff} = \frac{W_{wof}}{2A} \quad (1) \]

\[ K_{IC} = \frac{(2E'\gamma_{eff})^{1/2}}{} \quad (2) \]

where \( W_{wof} \) is the energy under L-D curve, and \( A \) is the area of ligament portion. \( E' \) is the elasticity for plane strain condition defined as \( E' = E/(1-\nu^2) \), where \( \nu \) is the Poisson's ratio. In the materials showing changing fracture resistance with crack extension, Eq. (2) yields the average value. Although the value of \( K_{IC} \) is correlated with \( \gamma_{eff} \) through \( E' \), \( K_{IC} \) dissipated its physical meaning at these temperatures, and was significant only in relative comparison as a substitute for \( \gamma_{eff} \).

The L-D curves of WOF tests and effective fracture energies were almost identical at 0.1 and 0.01 mm/min, but as the rate was lowered below 0.01 mm/min, effective fracture energy was increased with inflation of L-D curves. It reached the maximum value around 0.001 mm/min, which was almost triple of those above 0.01 mm/min. Then, the effective fracture energy began to decrease and resumed to show a less dependence on D-rate around 0.0002 mm/min.

As stated above, bridging or pulling out of grains are considered to be the primary factor for increasing fracture resistance of silicon nitride at high temperatures. The strong dependence of effective fracture energy on D-rate suggests the friction produced in pulling out of grains, which is affected by strain rate due to visco-elastic characteristics of glassy phase, makes great contribution into the change of effective fracture energy. The bridging effect of grains, particularly elongated ones, in the wake region following the crack tip is supposed to be a predominant factor for increase of effective fracture energy at 0.001 mm/min.

The change of effective fracture energy with D-rate may be explained as follows. In high rate region, or in quick stress application, the fracture occurs in brittle manner as seen from changes in both S-D curves and fractographies of tensile tests for similar silicon nitride at 1260°C, leading to low effective fracture energy. In this region, pulling out of grains is difficult to be produced because the interfacial shear stress at grain boundary is excessively high. As the D-rate is lowered, however, this stress is reduced to such a degree that the grains can be pulled out. In the range where the pulling out is possible, the higher shear stress yields larger fracture resistance, and hence effective fracture energy is decreased with the further decrease of D-rate.

Fracture resistance, \( K_R \), at a certain point of
The $L-D$ curve in WOF test can be calculated as follows:

$$K_R = (E'/2 \frac{d\varepsilon}{dA})^{1/2}P$$

(3)

where $\varepsilon$ is the compliance, $A$ is the crack area, and $P$ is the load. In CNB test fracture toughness is usually calculated from the maximum value of $P$ as $(E'/2 \frac{d\varepsilon}{dA})^{1/2}$ must take the minimum under the condition that $K_R$ is constant irrespective of crack length (flat $R$-curve behavior). The fracture toughnesses calculated from the maximum loads of 0.01 and 0.001 mm/min in Fig. 6 are 4.6 and 5.4 MPa m$^{1/2}$, respectively. This difference is much smaller than that between "average" values calculated using Eq. (2) as shown in Fig. 7, suggesting the increase of resistance with further extension of crack at 0.001 mm/min.

3.3 $R$-curve behavior and its $D$-rate dependence

By use of Eq. (3), the variations of fracture resistance, $K_R$, against crack growth, namely $R$-curve behaviors, were obtained. The results in WOF tests at 1260°C are shown in Fig. 8. Although WOF tests at 900°C and 1020°C resulted in flat $R$-curve behaviors which were similar to behaviors reported for silicon nitride by some researchers, rising $R$-curve behaviors were observed at 1260°C; particularly at 0.001 mm/min, $K_R$ grew almost double at 1260°C. Similarly to the variation of effective fracture energy with $D$-rate, $R$-curve showed the most intensive rising behavior around 0.001 mm/min.

Some toughening mechanisms are present for explaining rising $R$-curve behaviors for non-transformed ceramics; residual strain effects in process zone wake, grain bridging or grain interlocking effects, etc. Theoretical consideration indicated that $R$-curve showed steep increase of resistance in the beginning stage of crack extension, followed by "toughness plateau" corresponding to the stationary configuration of the advancing crack in a steady state. However, the experimentally obtained $R$-curve behavior is seriously affected not only by intrinsic material characteristics like grain size, etc., but also by experimental conditions such as depth of notch, specimen size, etc., and determination method of fracture resistance. In addition, crack branching and secondary crack formation make the definition of crack length ambiguous. The $R$-curve behaviors shown here involved not only the above uncertainties but also the unsteadiness of CNB test that crack width is varied with crack extension. Nevertheless, they are expedient because the bridging stress at crack interface can be estimated from the increment of fracture resistance, by assuming appropriate stress distribution as shown in the following section.

3.4 Bridging stress distribution in grain bridging zone

The bridging stress (or traction) at the crack interface in the grain bridging zone can be estimated from $R$-curves. The stress value can be simply defined when uniform bridging stress distribution in grain bridging zone is presumed as follows (Fig. 9, A):  

$$\sigma(X) = \sigma_0 \quad (X \leq X_0) \quad (4a)$$

$$\sigma(X) = 0 \quad (X > X_0) \quad (4b)$$

where $\sigma_0$ is the applied uniform stress, $X$ is the distance behind the crack tip, $a-x$ and $X_0$ is the bridging zone length. However, the uniform stress distribution is not likely to be produced, because this assumption needs singularities at $X = 0$ and $X_0$. The most probable stress distribution is described in Fig. 9, B; it increases from $\sigma(0) = 0$, reaches the maximum, and decreases tailing off to 0. In this study a cubic function was assumed to model this distribution as follows.

$$\sigma(X) = \sigma_1 X + \sigma_2 X^2 + \sigma_3 X^3 \quad (X \leq X_0) \quad (5a)$$

$$\sigma(X) = 0 \quad (X > X_0) \quad (5b)$$

where $\sigma_1$, $\sigma_2$, and $\sigma_3$ are coefficients. The continuity of function gives

$$\sigma_1 X_0 + \sigma_2 X_0^2 + \sigma_3 X_0^3 = 0 \quad (6)$$

Fig. 8. $R$-curve behaviors at 1260°C. Arrows indicate fracture toughness calculated from total WOF.

Fig. 9. Schematic of bridging stress distribution at crack interface. $a$ and $a_0$ are the crack lengths of extended and initial cracks, respectively.
The increment of resistance value, $\Delta K_s$, for the crack length, $a$, is expressed by

$$\Delta K_s = 2(a/\pi)^{1/2}(1/b(a)) \int_{a_0}^a \left[ \sigma(a-x) b(x)/(a^2-x^3)^{1/2} \right] dx \quad (7)$$

where $b(x) = b(x-a_0)/(W-a_0)$. By selecting $\sigma_0$, $\sigma_1$, $\sigma_2$, and $\sigma_3$ so that $R$-curve from Eq. (7) best fits with the experimentally obtained one, bridging stress distribution in bridging zone can be determined.

Figure 10 shows the bridging stress distributions obtained using Eqs. (5) and (6) for the case of 0.001 mm/min at 1260°C, and Fig. 11 shows $R$-curve calculated by assuming the stress distributions of Fig. 10 in comparison with the real $R$-curve. It is known that Eq. (6) owns a great superiority to Eq. (5) in expressing a real bridging stress distribution. The estimated bridging stresses, $\sigma_0$ and $\sigma_{\text{max}}$, are shown in comparison with tensile strength at 1260°C in the following section, but rather qualitative discussion should be permitted because the used $R$-curve themselves include some uncertainties, and $\sigma_{\text{max}}$ in Eq. (6) is largely changed by slight variation in the assumed shape of stress distribution. It seems possible, however, to think that actual bridging stress lies around them.

The crack opening displacement at the end of bridging zone can be determined for the uniform bridging zone as follows,

$$u = (8 \sigma_0 X_s/\pi E) + (8 \sigma_0 X_s^{1/2}/E'2\pi^{1/2}) \quad (8)$$

where $u$ is the total crack opening displacement, and $K_s$ is the toughness at the crack tip. The calculation for $\sigma_0$ of Fig. 10 resulted in $u=4 \mu$m.

As the length of elongated grain of hot-pressed silicon nitride is ranging from 5 to 10 $\mu$m, this displacement value seems plausible considering a bonding portion length between grain and matrix.

3.5 S-D diagrams in tensile tests

The above described grain bridging toughening mechanisms should have close relationship with the stress-strain curve, because pulling out of grains are caused by softening of glassy phase, which will bring a non-elastic stress-strain relation. The results of tensile tests at 1260°C are shown in Fig. 12, where the displacement was a real elongation of gauge section of tensile test specimen measured by electro-optical extensometer. As compared with the results of the similar tests in vacuum atmosphere, general behavior was almost the same. It is known that in some cases oxidation treatment decreases volume fraction of glassy phase and improves high temperature strength. In this study, however, almost no effect of oxidation was observed even for the case of 27 hours exposure at 1260°C.

While the test at 0.1 mm/min yielded almost linear $S-D$ curve up to fracture strength ($\sigma_1$), those at 0.05 mm/min and lower rates produced stress peaks ($\sigma_0$) after deviating from linearity,
and final strength ($\sigma_t$) at the rates from 0.005 to 0.0005 mm/min showed the convergence. Fractographic study revealed that fractures at 0.02 mm/min and lower rates exhibited "rough wake region", which was recognized as an evidence of "enhanced SCG" by several researchers, [1,2,40] while those at 0.1 mm/min and higher left no such a region, as shown in Fig. 13. This means the appearance of stress drop in S-D diagrams indicates an initiation of SCG.

Figure 14 shows the dependence of tensile strengths ($\sigma_t$, $\sigma_p$, and $\sigma_i$) and bridging stresses ($\sigma_b$ and $\sigma_{\text{max}}$) on D-rate. The bridging stress is raised with decreasing D-rate down to 0.001 mm/min at which the pulling out works of grains are most activated. The bridging stress is maximized at 0.001 mm/min like effective fracture energy. It is known that the appearance of SCG wake region in fracture surface of silicon nitride accompanies the steep rise of fracture toughness. [1,13]-15,19] Thus, it can be supposed that the maxima of bridging stress and effective fracture energy at 0.001 mm/min are correspond-

![Fig. 13. Optical images of fracture surfaces tested at (a) 0.1 and (b) 0.02 mm/min. Arrows indicate "enhanced" SCG wake.](image)

Fig. 14. D-rate dependences of bridging stresses and tensile strengths.

The plastic behavior (yielding phenomena) observed in tensile tests at some low D-rates as shown in Fig. 12 is attributed to slow crack propagation accelerated by spread cavities. This crack growth is accompanied by pulling out works of grains, which should be a primary factor for increasing fracture resistance and make a great contribution for bearing applied stress while crack propagates. As the uniform stress is applied in a tensile test, the cracks are produced everywhere throughout a gauge part of specimen. This crack is supposed to be slowly propagated up to the length of about 10 $\mu$m and then be arrested by grains elongated in the stress direction, because 10 $\mu$m is the average interval of elongated grains. [38] In the process that this medium-sized crack grows to macrocrack by connecting each other, pulling out of elongated grains should be generated. Thus, the bridging of grains at the interface of this slowly growing crack is assumed to be the same in its mechanism as that in CNB test. As a matter of fact, the comparison in Fig. 14 revealed that the bridging stress estimated from R-curve behaviors in WOF tests corresponded to a fairly large part of the tensile strength.

4. Conclusions

Chevron notch bend test of hot-pressed silicon nitride at high temperatures above 1000°C demon-
strained that the characteristics concerning fracture resistance were largely dependent on \( D \)-rate; at a certain \( D \)-rate (0.001 \text{mm/min} in this study) effective fracture energy took the maximum value and \( R \)-curve showed the strongest rising behavior. It was assumed that an increase in fracture resistance and a strong \( R \)-curve behavior were attributed to activated pulling out works of grains, which were heavily affected by a rate-sensitive grain boundary shear stress. The bridging stress at crack interface, which was estimated from the increment of fracture resistance with crack extension, exhibited the \( D \)-rate dependence very similar to that of effective fracture energy. The maximum bridging stress was supposed corresponding to a tensile strength showing stress drop in its \( S-D \) curve, from an appearance of SCG wake region. In \( D \)-rate range below the rates giving this correspondence, tensile strength and bridging stress presented the similar \( D \)-rate dependence. The bridging stress corresponded to a fairly large part of the tensile strength, and it was implied that, in high temperature tensile test, the bridging of silicon nitride elongated grains should make a great contribution for bearing an applied tensile stress during slow crack growth.

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References

28) T. Ohji, unpublished work.