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Fracture of silicon nitride joined with an aluminium braze

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Mechanical Properties of silicon nitride joined with aluminium braze have been investigated using fracture mechanics. The highest bonding temperature, 1133 K, produced the highest four-point bend strength of 417 MPa, the strength depending strongly on stress rate. The fracture parameter, N, for slow crack growth in the joint was 29.7 which was near to that of the silicon nitride. Stress corrosion cracking is believed to be one of the serious problems associated with ceramic joining.

1. Introduction

As silicon nitride has several more useful properties than traditional oxide ceramics, such as high-temperature strength, wear resistance, etc., the development of practical applications has further increased its importance. Metal/ceramic and ceramic/ceramic joining techniques are thought to be of key importance for the practical use of ceramics as structural materials. For successful joining of ceramics to metals, the problem of residual stress resulting from different thermal expansion coefficients between these materials must first be resolved. In joining ceramics of the same type, where a joining layer is very thin, the problem of residual stress is not so critical.

There are several methods for ceramic/ceramic joining. For example, a solid-state bonding method using refractory metals such as tungsten, molybdenum, tantalum [1], a direct bonding method using high pressure [2], an active metal brazing method [3], a glassy solder method [4] and a ZrO_2-adhesive method [5]. Among these methods, the active metal brazing method is a very simple process and the joints obtained have relatively high strength. Recently, two of the present authors have discovered that silicon nitride joints using pure aluminium solder have a very thin joining layer and high joining strength which is nearly the same as that of silicon nitride [6, 7]. However, it is generally known that the strength of brittle materials such as ceramics depends on stress rate in some environments [8, 9], i.e. slow crack growth (SCG) occurs via stress corrosion. Therefore, when ceramic joints are used in structural materials, it is very important to know the fracture mechanics parameters which relate to SCG. The aim of the present work is to investigate such parameters concerned with fracture of silicon nitride joints.

2. Experimental procedure

2.1. Preparation of materials and bonding

Pressureless-sintered silicon nitride with 5 wt % $MgAl_2O_4$ and 5 wt % Y_2O_3 as additives used in the present work was supplied as rods and blocks. Rods were used to investigate the dependence of strength on joining temperature and interfacial structure. Surfaces of rods 5 mm diameter and 20 mm long were ground with a no. 450 diamond wheel to a surface roughness of 0.4 μm R_z (ten points average surface roughness standardized in JIS B0601)*. Pure aluminium foil (JIS A1050) 6 mm diameter and 0.1 mm thick was placed between two silicon nitride rods in a BN mould.

The apparatus used for ceramic bonding is illustrated in Fig. 1. The BN mould was set in the centre of a hot-pressing chamber which was evacuated to 5×10^{-6} torr. Bonding pressure was applied on the specimen equivalent to a uni-axial compressive stress of 3.3 MPa. The specimen was heated to a bonding temperature in the range 933 to 1133 K and held at that temperature for 15 min.

Blocks of silicon nitride were used to investigate the stress-rate dependence of fracture strength of joints and their fracture toughness (K_{IC}). The dimensions of a bond face were 10 mm × 19 mm and the face was ground by the method described above. Pure aluminium foil was placed between two silicon nitride blocks which were joined under a uni-axial compressive stress of 0.3 MPa at 1133 K for 15 min in a vacuum of 5×10^{-6} torr.

2.2. Bending tests

After joining, rods were directly used as bending specimens and block joints were cut and ground to a test bar of 3 mm by 3 mm by 38 mm. Specimens for measuring K_{IC} had a single edge notch of 100 μm wide

*$R_z = 1/10(R_{+1} + R_{+2} + \cdots + R_{+5} + R_{-1} + R_{-2} + \cdots + R_{-5})$, where R_{+1} is the highest peak height, R_{+2} the second highest, etc., R_{-1} is the deepest scratch depth, R_{-2} the second deepest, etc.

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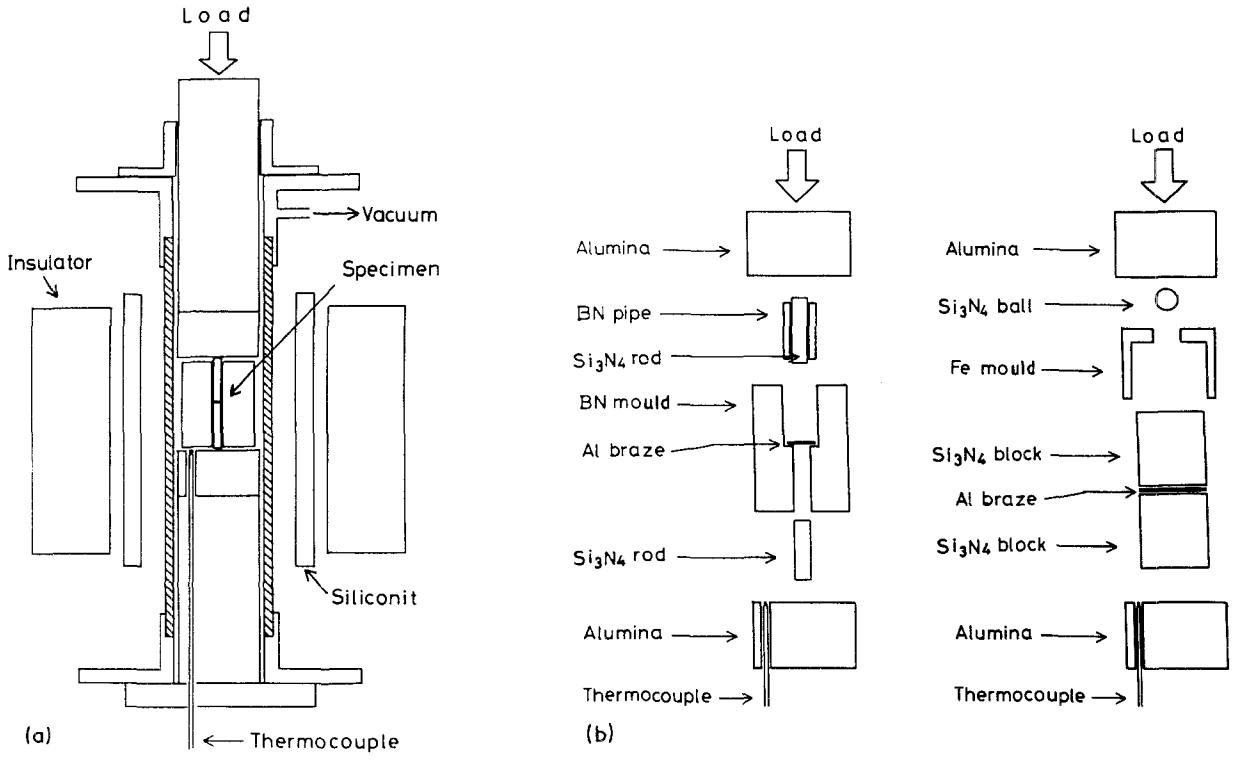

Figure 1 (a) A schematic drawing of the vacuum hot-pressing apparatus. (b) Assembly for bonding.

and 1 mm deep which was introduced at the position of the joining layer with a diamond blade 100 μm thick. Four-point bending tests (upper span 10 mm, lower span 30 mm) were performed using an Instron testing machine at room temperature mainly in air, partly in water, and in dry argon. The cross-head speed was 0.5 mm min^{-1} but in the case of stress rate tests was varied in the range 0.16 to 220 mm min^{-1}. To measure the fracture load precisely at high stress rate, a transient memory scope was used.

2.3. Microstructural analysis

Element distribution across a joining layer was examined by X-ray microanalysis (XMA) and an X-ray diffraction method (XRD) was used to identify reaction products on fracture surfaces. After the bending test, the observation of fracture surfaces was performed by scanning electron microscopy (SEM).

3. Results and discussion

3.1. Microstructure and effect of bonding temperature on strength

Fig. 2 shows the microstructure of a silicon nitride/aluminium/silicon nitride interface. The aluminium brazing layer was less than 10 μm thick. No reaction layer was observed on the silicon nitride/aluminium interface determined by XMA or by XRD of the fracture surface. Recent work reported the formation of 15R AlN type sialon layer and amorphous alumina layer at the silicon nitride/aluminium interface reacted at 1473 K [10]. The present authors also recognized two reaction layers (β'-sialon and Al–Si–O amorphous layers) between HIPed silicon nitride without additive and aluminium joined at 1073 K [11]. The total thickness was thinner than 500 nm. SEM, XMA or XRD, which do not have enough resolution, could not distinguish them.

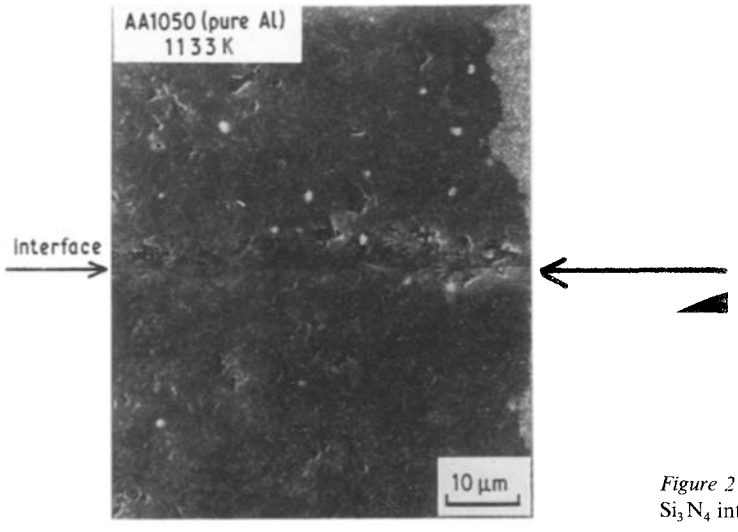

Figure 2 A scanning electron micrograph of Si_3N_4/aluminium/Si_3N_4 interface.

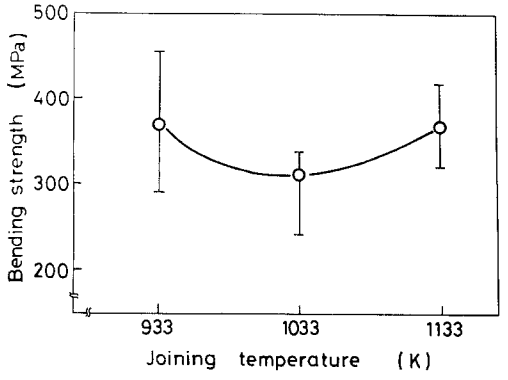

Figure 3 Effect of bonding temperature on bending strength.

Fig. 3 shows the bonding-temperature dependence of bending strength. With increasing bonding temperature the mean strength shows a decrease between 933 and 1133 K, with further scatter in strength at the lower bonding temperature. The maximum bending strength was obtained by bonding at 933 K. The fracture surfaces of the joints formed at 1033 K show many unbonded areas (Fig. 4) which were absent in the surfaces joined at 933 K. Because the temperature of 933 K is just below the melting point of pure aluminium, the plasticity by viscous flow of pure aluminium at 1033 K is higher than that by plastic flow at 933 K. Furthermore, it is well known that aluminium does not wet silicon nitride [12]. Hence pure aluminium would flow out from the interface of two rods of silicon nitride bonded at 1033 K and result in unjoined areas.

K_{IC} was measured on the joint bonded at 1133 K and was 5.5 MPa $m^{1/2}$ on average. This value is almost the same as that of the silicon nitride used in the present study.

3.2. Stress-rate dependence of fracture strength

It is well known that, when water is present in the testing atmosphere, a crack in ceramics can propagate below the critical stress for instantaneous fracture in an inert atmosphere. This phenomenon is sometimes called "fatigue" [9, 13], and may be expressed as an exponential relationship between crack velocity (V) and stress intensity factor (K_I)

$$V = AK_I^N \quad (1)$$

where N and A are constants determined by the type of material and environment and are parameters used to describe slow crack growth. When stress is applied at a constant stress-rate to a specimen, applied stress at a time t is

$$\sigma(t) = \dot{\sigma}t (\dot{\sigma} = \text{const.}) \quad (2)$$

Assuming that σ_i is an inert strength of one surface crack which determine the strength of a specimen and that σ_f is some environmental strength determined by the same crack, the relationship between σ_i and σ_f is derived from Equations 1 and 2 [7, 14].

$$\sigma_f^{N+1} = \left(\frac{N+1}{N-2}\right)\frac{2\dot{\sigma}}{AY^2}\left(\frac{\sigma_i}{K_{IC}}\right)^{N-2} \quad (3)$$

where Y is a constant determined by notch geometry and applied condition of stress and K_{IC} is fracture toughness. Taking logarithms of both sides of Equation 3 leads to

$$\log \sigma_f = \frac{1}{N+1}\log \dot{\sigma} + C \quad (4)$$

where

$$C = \frac{1}{N+1}\log\left[\left(\frac{N+1}{N-2}\right)\frac{2}{AY^2}\left(\frac{\sigma_i}{K_{IC}}\right)^{N-2}\right]$$

From Equation 4, a plot of log σ_f against log $\dot{\sigma}$ should be given as a straight line with slope of $1/(N+1)$, from which the value of N can be calculated.

Fig. 5 shows the log σ_f–log $\dot{\sigma}$ plot of the silicon nitride joined with aluminium. Bending tests were performed in air of 60% humidity at room temperature. Open circles in this figure denote the arithmetical average of four or six measured values. From this figure, the scatter of fracture strength decreases with increasing stress rate. Assuming a linear relationship between fracture strength and stress rate, the value of N was calculated to be 29.7 using the least squares method from the plot. The fracture path changed with stress rate as shown in Fig. 6. The fracture origin was located near the interface in all cases except at the highest stress rate. The fracture paths often propagate within the silicon nitride near the interface with increasing stress rate, compared to interfacial (ceramic/Al) fracture with decreasing stress rate. The flaws which initiate to fracture in the low-strength specimens are sometimes unjoined areas at the interface. At higher stress rates, they are often in the vicinity of the interface. The fracture propagated in silicon

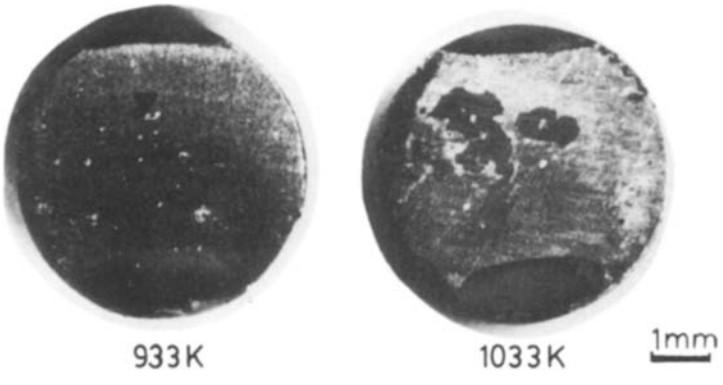

Figure 4 Effect of bonding temperature on the appearance of the fracture surface.

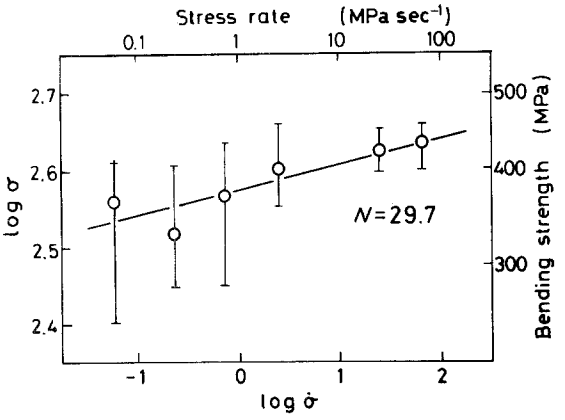

Figure 5 Stress-rate dependence of fracture stress.

nitride near the interface (Fig. 7). Fig. 8 shows the microstructure of aluminium braze which remained on the fracture surface, and it is observed that the fracture surface consisted of shear fracture of aluminium braze and brittle fracture of silicon nitride in the case of slow stress rates, but at high stress rates it was a mixed structure of dimple-like fracture structure within aluminium braze, which indicates ductile fracture, in addition to the structure described above.

In order to determine what controls the fracture of the joint and to compare the mechanical properties of the joint with those of silicon nitride itself, the stress-rate dependence of fracture strength of the silicon nitride was measured, and is summarized in Fig. 9. In the range of $\dot{\sigma} < 2.7\,\mathrm{MPa\,sec^{-1}}$, fracture strength showed a stress-rate dependence with a value of N of 27.5, but when $\dot{\sigma} > 2.7\,\mathrm{MPa\,sec^{-1}}$, fracture strength became constant.

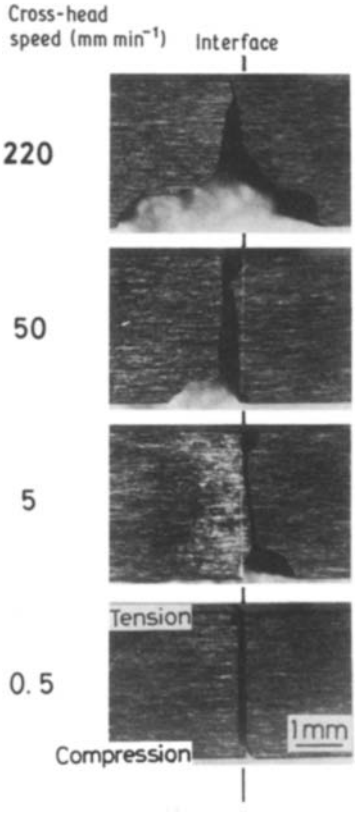

Figure 6 Macrographs showing the difference of the fracture path in bending specimens at various stress rates.

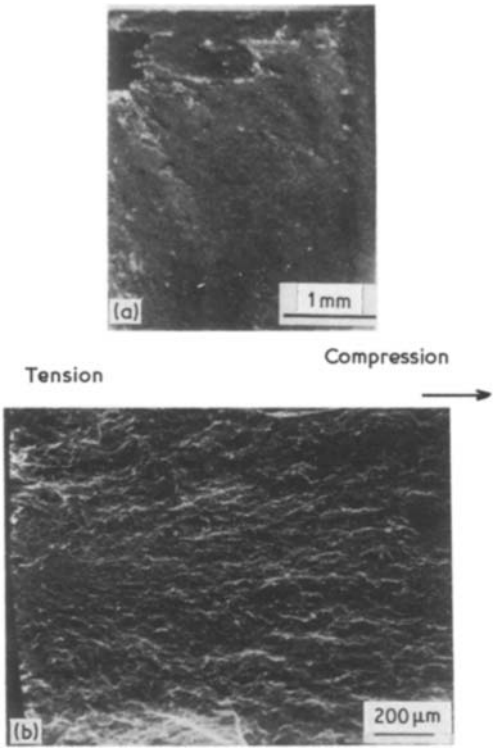

Figure 7 Scanning electron micrographs of flaws connected to fracture; (a) unjointed areas and (b) micropore and fracture paths.

Two important facts were obtained from the discussion above; (i) the value of N of the joint was close to that of silicon nitride, and (ii) at high stress rates the fracture stress of the joint was nearly the same as that of silicon nitride and the fracture in the joint often propagated deeply in the silicon nitride. From facts (i) and (ii), the fracture of the joint seems to be controlled by the mechanical properties of silicon nitride itself and the stress-rate dependence indicates the SCG phenomenon in the joint.

In order to confirm these results, bending tests were performed in different environments. The results are summarized in Table I. Because humidity in air is rather high (73% humidity), the bending strength in air was not very different from that in water. But comparison of the data in water and in dry argon showed a difference of more than 100 MPa in the bending strength. The difference might be caused by the stress corrosion, near the interface within the silicon nitride, by water. Hence, it is concluded that silicon nitride controls the fracture of the joint. The correspondence of K_{IC} for the joint and the silicon nitride also supports this conclusion. In addition, although the inert strength of the joint reached that of silicon nitride, the joint strength in a moist

TABLE I The bending strength of the joint in several environments

	Water (MPa)	Air (MPa) (73% humidity)	Dried argon (MPa)
Specimen 1	335.5	352.8	435.6
2	337.2	356.5	455.2
3	398.5	407.2	508.5
Average	357.1	372.2	466.4

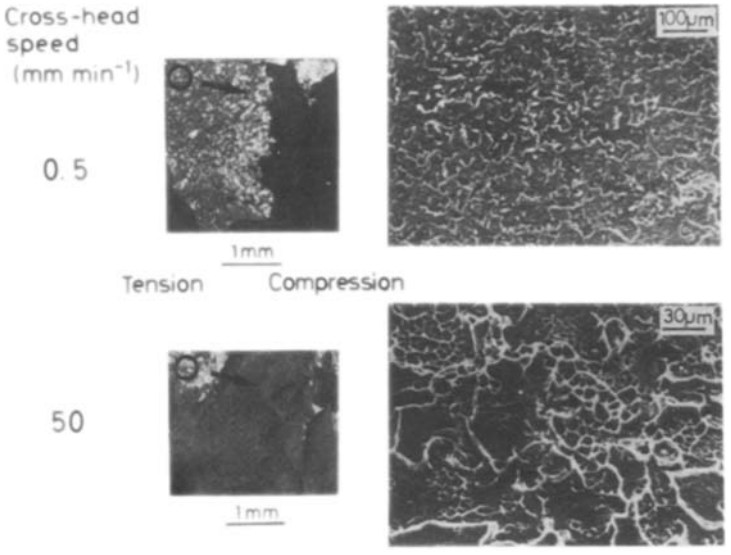

Figure 8 Microstructures of aluminium braze which was left on fracture surface.

atmosphere was lower than that of silicon nitride. This fact suggests that the SCG phenomenon would degrade the joint much more than the silicon nitride.

Ceramic/ceramic or ceramic/metal joints always have defects on the interface, such as grinding scratches and deep valleys produced from a bonding reaction. In such cases, SCG will be an additional problem in applying the joint to highly stressed structural components, because the interface with those defects will be severely attacked.

4. Conclusion

Silicon nitride was joined using an aluminium braze and the mechanical properties of the joint were investigated. Some of the joints approached the bending strength of the silicon nitride. Therefore, aluminium is confirmed as a good brazing material for silicon nitride.

The strength of joints shows a remarkable dependence of stress rate and the parameter of crack propagation, i.e. N value, is 29.7, similar to that for silicon nitride (27.5). The strength of the joint was much lower in water than in dry argon. It is believed that a crack propagates in silicon nitride near the interface due to stress corrosion by water. Fracture of the joint is mainly controlled by the properties of the silicon nitride influenced by the brazing reaction and additional interfacial flaws in the silicon nitride side.

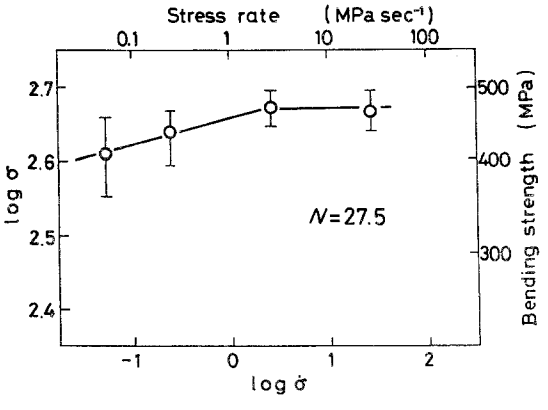

Figure 9 Stress-rate dependence of fracture stress of silicon nitride specimens.

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