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Joining of silicon nitride ceramics by hot pressing

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The joining of hot-pressed silicon nitride ceramics, containing Al_2O_3 and Y_2O_3 as sintering aids, has been carried out in a nitrogen atmosphere. Uniaxial pressure was applied at high temperature during the joining process. Polyethylene was used as a joining agent. Joining strength was measured by four-point bending tests. The effects of joining conditions such as temperature (from 1400 to 1600° C), joining pressure (from 0.1 to 40 MPa), holding time (from 0.5 to 8 h) and surface roughness (R_{max}) of the joining couple (about 0.12, 0.22 and 1.2 μm) on the joining strength were examined. The joining strength was increased with increases in joining temperature, joining pressure and holding time. Larger surface roughness caused lower joining strength. The higher joining strength was attributed to a larger true contact area. The area was increased through plastic deformation of the joined couple at elevated temperatures. The highest joining strength attained was 567 MPa at room temperature, which was about half the value of the average flexural strength of the original body. The high temperature strength measured at 1200° C did not differ very much from the room-temperature value.

1. Introduction

Silicon nitride ceramics have been considered as one of the most promising structural materials for high temperature applications. However, the difficulty in the fabrication process of complex-shaped and larger-sized components requires the development of a joining technology for ceramics.

So far, several attempts have been carried out for the joining of silicon nitride ceramics with the use of some joining agents such as ZrO_2 [1], a mixture of SiO_2–CaO–MgO–Si_3N_4 [2], a mixture of Y_2O_3–La_2O_3–MgO–Si_3N_4 [3], an MgO–Al_2O_3–SiO_2 glass composition [4] and kaolin with some fluoride [5]. However, the presence of these foreign materials results in the formation of glassy phases at the joining interface and consequently causes a degradation of the high-temperature strength of the joined parts.

Recently, joining processes of silicon nitride ceramics by hot isostatic pressing [6], uniaxial hot pressing [7] and by reaction bonding [8] without any extra materials on the joining interface have been reported. In these cases, the joined components are expected to maintain the same strength as the original body at high temperatures even above 1200° C. However, the effect of the joining conditions on the joining strength has not been established yet.

In the present study, joining of silicon nitride ceramics with the use of polyethylene film as a joining agent was carried out in a hot-pressing furnace in a nitrogen atmosphere with varied joining conditions such as temperature, pressure and holding time. The joining strength of the specimens was measured by four-point bending tests on test pieces prepared from the joined specimens. The relationships between the joining strength and the joining conditions are discussed.

2. Experimental procedure

Commercially available hot-pressed silicon nitride ceramics (Toshiba Corporation) were used as specimens to be joined. Some characterized properties of the ceramics are shown in Table I.

The as-obtained ceramic body was cut into pieces of size about 15 mm × 15 mm × 19 mm. Most of the specimen surfaces to be joined were ground with a 400 grit diamond grinding wheel. Their surface roughness (R_{max}) was about 1.2 μm. Some of the pieces were polished with a 1000 or 400 grit diamond disc in order to prepare much smoother surfaces of about 0.12 and 0.22 μm, respectively.

Two pieces of the specimens were attached together as a joining couple. A polyethylene sheet of thickness 40 μm was put between the two pieces, which were stuck together by melting the sheet in a vacuum at about 170° C before the joining operation. During the heating process in a nitrogen atmosphere, the polyethylene sheet is expected to leave a certain amount of carbon, which would react with silicon nitride to form

TABLE I Some properties of hot-pressed Si_3N_4

Amount of additives (wt %)		Bulk density ($g\,cm^{-3}$)	Flexural strength (GPa)	Weibull's modulus
Al_2O_3	Y_2O_3			
2.9	5.0	3.196	1.05	8.2

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TABLE II Joining conditions

Temperature	1400 to 1600° C
Pressure	0.1 to 40 MPa
Holding time	0.5 to 8.0 h
Joining agent	None or polyethylene (thickness 40 μm)
Atmosphere	1 atm N_2

TABLE III Conditions of four-point bend test

Upper span	10.0 mm
Lower span	20.0* or 30.0† mm
Loading rate	0.5 mm min^{-1}
Radius of span edge	2.5 mm

*Joining strength.
†Flexural strength.

a silicon carbide layer between the joined specimens according to the reaction

$$Si_3N_4 + 3C \rightarrow 3SiC + 2N_2$$

The formation of the silicon carbide was expected to increase the joining strength.

The attached couple was then inserted into a graphite mould. In order to prevent the reaction between silicon nitride and graphite, thin boron nitride plates were put between the graphite punches and the joining couple. The open space in the mould was also stuffed with boron nitride powder.

The graphite mould was placed into an induction heating hot-pressing furnace. The joining conditions are shown in Table II. The mould was heated up to the joining temperature with a heating rate of 25° C min^{-1}.

After the joining process was performed, the joined couples were cut perpendicular to the joining interfaces to prepare test pieces for joining-strength measurement. The side faces of the test pieces were ground with 200 and 400 grit resin-bonded diamond grinding wheels in turn. Then the test pieces were chamfered with a 600 grit diamond disc. Twelve pieces of size 3 mm × 4 mm × 30 mm were prepared from each joined couple. The joining strength was measured by the four-point bending method, with the testing conditions shown in Table III.

Fracture surfaces of the joined specimens and joining interfaces were examined by a scanning electron microscope (SEM) and a transmission electron microscope (TEM) equipped with an energy-dispersive X-ray spectroscope (EDX).

In order to examine the effect of annealing on the strength of the ceramic bodies used, blocks of size 30 mm × 40 mm × 15 mm were heated at temperatures between 1400 and 1700° C in 1 atm nitrogen atmosphere for 2 h. The annealed blocks were cut into fifteen test pieces of size 3 mm × 4 mm × 40 mm. The flexural strength was measured at room temperature with the test condition shown in Table III.

3. Results and discussion

3.1. Plastic deformation during joining process

Deformations of the specimens were recognized after the joining process, especially when pressure joining was carried out. The relative reduction of the specimen length is shown in Fig. 1. With a joining pressure of 20 MPa the deformation was remarkable at 1600° C, while with a pressure of 0.1 MPa or at temperatures below 1500° C the deformation was almost negligible. Therefore, when joined above 1500° C, the pressure should be kept as low as possible in order to prevent the detrimental deformation of the joined component.

3.2. Effect of joining temperature

Fig. 2 shows the effects of joining temperature and polyethylene as a joining agent on the joining strength, when joined with a joining pressure of 20 MPa. It is obvious that the joining strength was increased with an increase in joining temperature, and the use of polyethylene sheet also increased the joining strength.

Scanning electron micrographs of the joining interfaces are shown in Fig. 3. The real contact area joined at 1600° C was apparently larger than that joined at 1400° C. This can be explained by the faster creep deformation rate of the silicon nitride ceramics containing glassy grain-boundary phases at higher temperatures.

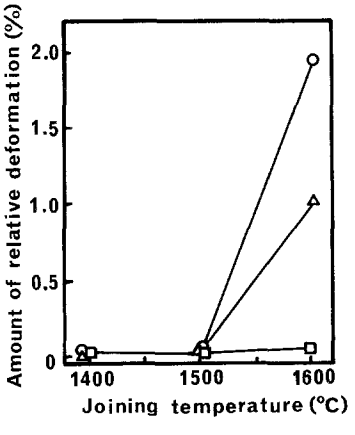

Figure 1 Amount of relative deformation of joined specimen. Initial height of joined specimen: H_0 = 30 mm. Pressure and holding time: (○) 20 MPa, 1.0 h; (△) 20 MPa, 0.5 h; (□) 0.1 MPa, 0.5 h.

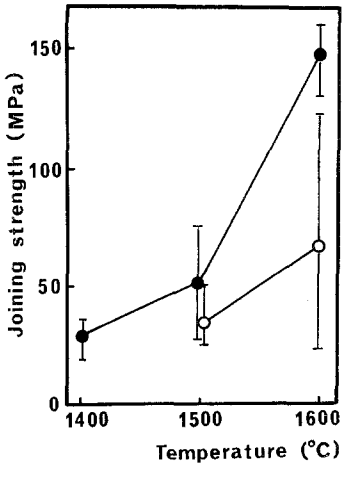

Figure 2 Effect of joining temperature on joining strength. (●) With polyethylene, (○) without polyethylene. Holding time: 0.5 h, joining pressure: 20 MPa.

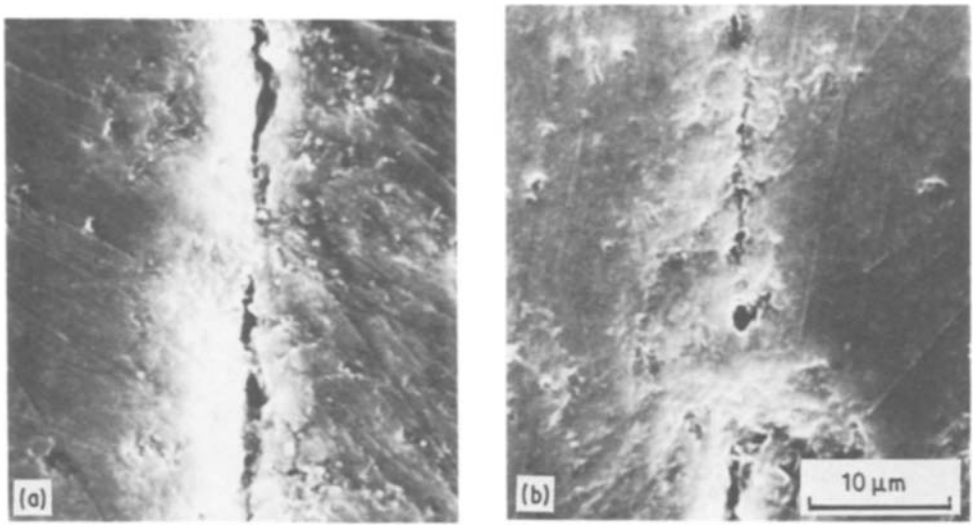

Figure 3 Joining interfaces. Joining temperature: (a) 1400° C, (b) 1600°C. Joining pressure: 20 MPa, holding time: 1 h, joining with polyethylene.

Fig. 4 shows the effect of heat treatment of the hot-pressed silicon nitride bodies on their room-temperature flexural strength. The flexural strength decreased gradually with an increase in the heat treatment temperature. When the specimen was heated at 1700° C for 2 h, the average strength was reduced from 1050 MPa to about 800 MPa. Even though the flexural strength of the annealed body was degraded to some extent, the strength value of the annealed body is still appreciably higher than the joining strength. Therefore, the overall mechanical strength of the joined specimens was improved as the joining temperature increased.

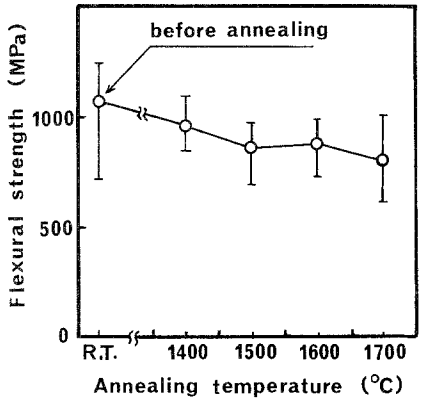

Figure 4 Effect of annealing on room-temperature strength of hot-pressed silicon nitride ceramics with Y_2O_3–Al_2O_3 additives. Annealing conditions: 1 atm N_2, holding time: 2 h. R.T. = Room temperature.

3.3. Joining pressure

Fig. 5 shows the effect of joining pressure on the joining strength. When the joining temperature was 1500° C, the joining strength was increased only slightly by an increase in joining pressure. When joining was carried out at 1600° C, however, the joining strength increased remarkably with an increase in pressure. This tendency corresponds well to the deformation behaviour of specimens shown in Fig. 1. Consequently, it is considered that some degree of deformation in the region near the joining interface is necessary to expand the true contact area and accordingly increase the joining strength with this method.

3.4. Holding time

Fig. 6 shows the effects of holding time on the joining

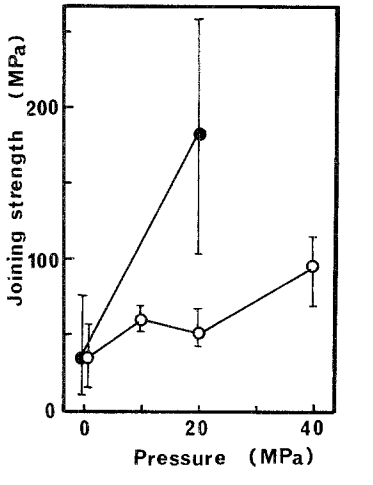

Figure 5 Effect of joining pressure on joining strength. Joining temperature: (○) 1500° C, (●) 1600° C. Holding time: 1 h.

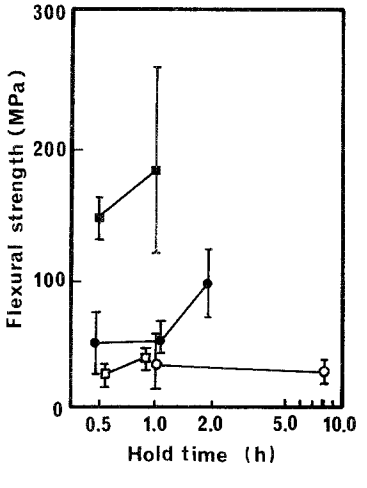

Figure 6 Effect of holding time on joining strength. Joining temperature and prssure: (□) 1400° C, 20 MPa; (○) 1500° C, 0.1 MPa; (●) 1500° C, 20 MPa; (■) 1600° C, 20 MPa.

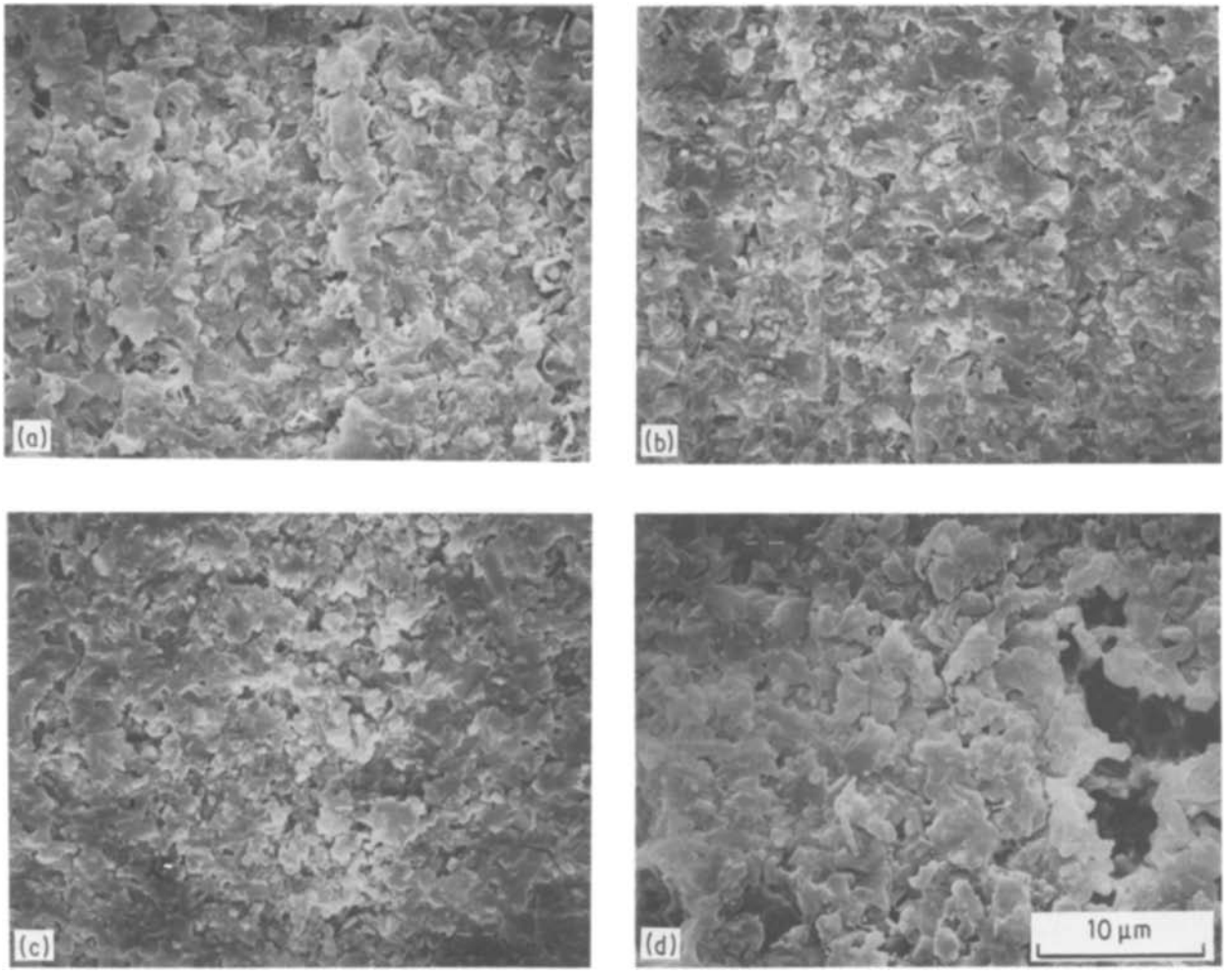

Figure 7 Porous interfacial zones exposed on fracturing. Joining temperature: 1500° C, joining pressure: 0.1 MPa, holding time: (a) 1 h, (b) 2 h, (c) 4 h, (d) 8 h.

strength. With a pressure of 0.1 MPa at 1500° C, an extended holding time was not effective to increase the joining strength. However, with a pressure of 20 MPa, an increase in joining strength with an increase of holding time was observed.

Fracture surfaces of the joined specimens are shown in Fig. 7. With a holding time of 8 h at 1500° C, relatively large open pores and a porous zone were observed on the fracture surface. The formation of these pores during the heating period is considered to cause the reduction of joining strength.

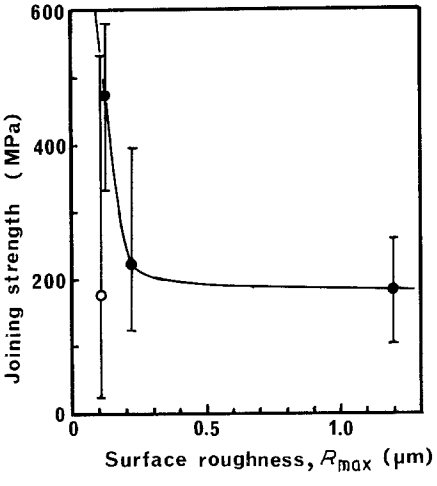

Figure 8 Effect of surface roughness on joining strength. Joining pressure: (●) 20 MPa, (○) 0.1 MPa. Joining temperature: 1600° C, holding time 1 h.

3.5. Roughness of joining surface

Fig. 8 shows the effect of surface roughness of the joining couples on the joining strength. With a joining pressure of 20 MPa, a small degree of roughness increased the joining strength remarkably. The maximum joining strength reached 567 MPa, which was about half the average strength of the original body.

However, with a pressure of 0.1 MPa, even though the surface roughness was small, the average joining strength was fairly low and the strength values were scattered widely. This scattering is considered to be due to the very slight curvature of the surfaces, which reduces the true contact area between the joining specimens. Then, as shown in Fig. 9, many more closed pores remained in the interfacial zone joined with a pressure of 0.1 MPa than with the pressure of 20.MPa. It was therefore concluded that without some joining pressure it is very difficult to achieve reliable joining with this process.

3.6. TEM and EDX analysis

A TEM photograph of the joining interface is shown in Fig. 10. The average grain size in the joining zone was slightly smaller than that in other regions.

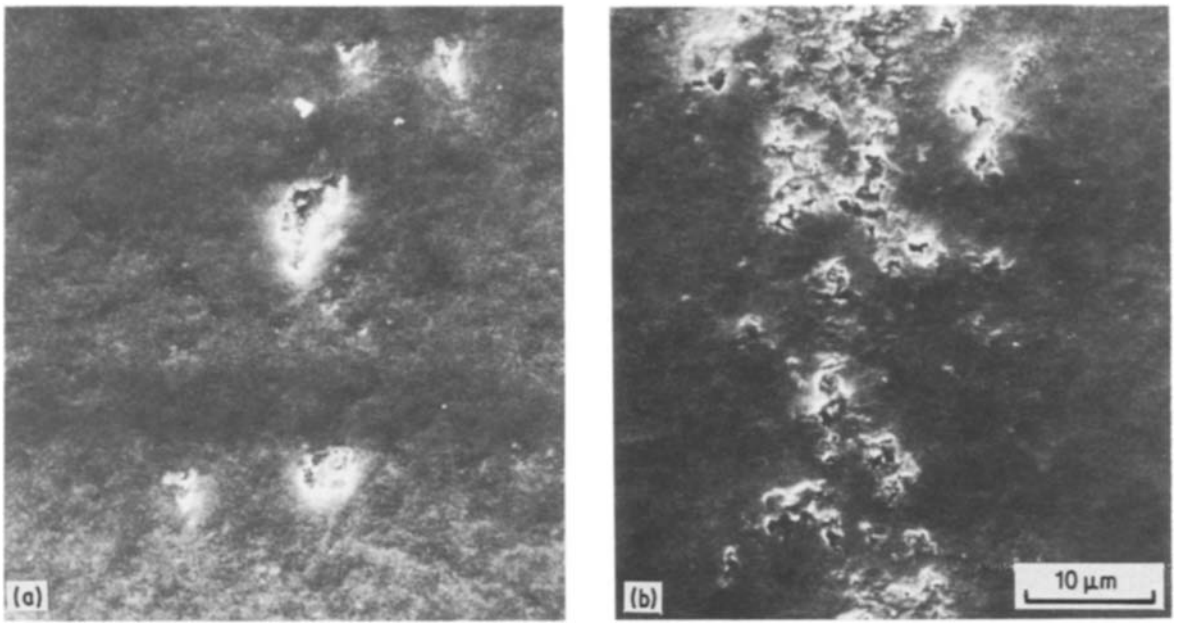

Figure 9 Closed pores at joining interfaces. Joining pressure: (a) 20 MPa, (b) 0.1 MPa. Holding time: 1 h, joining temperature: 1600° C, surface roughness (R_{max}): 0.12 μm.

Fig. 11 shows the results of EDX analysis. Although the formation of silicon carbide by the reaction between silicon nitride and residual carbon was expected, no appreciable difference in chemical composition between the joining zone and the original body was recognized.

3.7. Joining strength at high temperature

Fig. 12 shows the temperature dependence of the strength of joined specimens. It was recognized that the joining strength values at elevated temperatures were almost the same as the room-temperature strength, in spite of the evident decrease in the flexural strength of the original body above 1000° C.

4. Conclusions

The effects of the joining conditions of hot-pressed silicon nitride ceramics containing Al_2O_3 and Y_2O_3 as sintering aids on the joining strength have been examined. The joining strength was increased with increases in joining temperature, joining pressure and

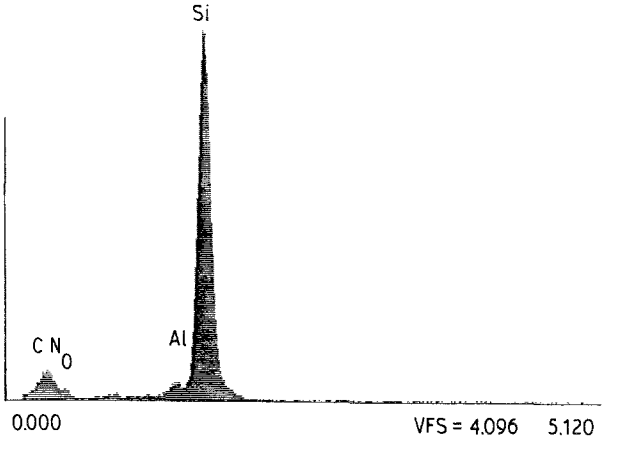

Figure 11 X-ray energy spectrum from the joining interface.

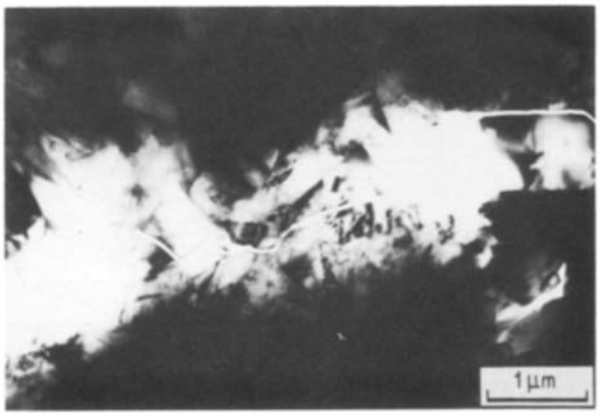

Figure 10 Transmission electron micrograph of joining interface. Joining temperature: 1600° C, joining pressure: 20 MPa, holding time: 1 h, surface roughness (R_{max}): 0.12 μm.

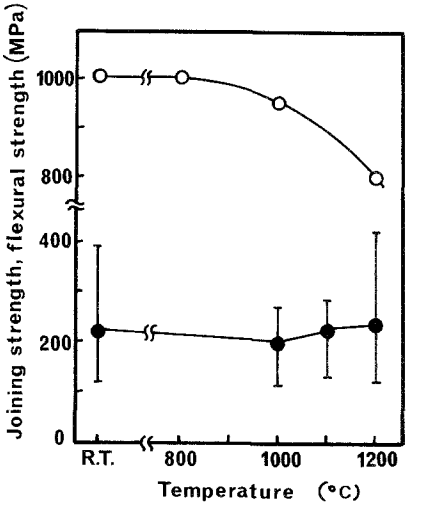

Figure 12 Joining strength at high temperature. (●) Joining strength, (○) flexural strength of HP-Si_3N_4. Joining temperature: 1600° C, Joining pressure: 20 MPa, holding time: 1 h, surface roughness (R_{max}): 0.22 μm.

holding time. However, a decrease in the surface roughness of the specimens increased the joining strength noticeably. The increase in joining strength was attributed to an enlargement of the true contact area, which was increased by creep deformation near the joining interface. Therefore, in order to increase the joining strength, it was necessary to reduce the surface roughness of the specimens to be joined.

Furthermore, the joining strength maintained almost the same value at temperatures up to 1200° C in air. Silicon nitride ceramic components joined with this process could be used at elevated temperatures as high as those used for bulk silicon nitride ceramics.

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