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Ceramics International 25 (1999) 101–105

CERAMICS
INTERNATIONAL

Effects of chemical compositions of adhesive and joining processes on bond strength of Si_3N_4/Si_3N_4 joints

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Received 3 October 1997; accepted 8 January 1998

Abstract

Silicon nitride ceramics were pressurelessly joined using glass adhesives with different α-Si_3N_4 powder additions at the temperatures ranging from 1450 to 1750°C for 10–120 min. The bond strength of the joint decreased sharply with increasing α-Si_3N_4 content in the adhesive. An optimum strength, 400 MPa, was obtained when joining was performed at 1650°C for 30 min. The results of the microstructural analysis showed that an intimate and glass-filled bond was produced where thermal cracks were also found clearly. © 1999 Elsevier Science Limited and Techna S.r.l. All rights reserved

Keywords: A. Joining; B. Microstructure; D. Silicon nitrate; Bend strength

1. Introduction

Structural and other applications of silicon nitride (Si_3N_4) have been expected because of its good high-temperature strength, wear resistance, and anticorrosion characteristics. But there are many cases in which the technology to bond Si_3N_4 to itself or to metals is required for practical applications. Brazing and diffusion bonding are the most promising methods for practical applications. The filler materials may be metals or alloys [1–6]. Although high bond strength can be obtained by using metal interlayers, the microstructures of the bonded region are quite different from those of the ceramics bonded, which can introduce some thermal cracks in the joint due to the thermal expansion coefficient mismatch between them. Meanwhile, the joint with different microstructures can not exhibit good chemical attack as well as the base materials.

It is well known that sintering aids are used in the densification of silicon nitride ceramics, these sintering aids usually react with the Si_3N_4 but still remain in the final microstructure as glassy grain boundary phases. It has been recognized for some time that glasses, particularly those used to assist densification during sintering, also have the potential to be used for joining Si_3N_4 ceramics. There are two advantages of using glass adhesive as filler materials for joining [7]: (a) chemical compatibility with Si_3N_4 ceramic is generally assured; (b) some physical properties such as viscosity, flow properties, and melting characteristics of glasses can be controlled over wide ranges. In addition, glasses have superior corrosion resistance to metallic filler materials. With this in mind, various workers have had successfully joining Si_3N_4 ceramics using glass solder [8–12].

In the present study, we pressurelessly join Si_3N_4 ceramics to itself using glass adhesives in the system Y_2O_3–Al_2O_3–SiO_2–Si_3N_4. Three adhesives are designed with different α-Si_3N_4 contents in order to investigate the effect of the chemical composition of adhesives on the joint strength. At the same time, the joining conditions such as temperature, hold time are examined.

2. Experimental procedures

2.1. Materials

The pressurelessly sintered Si_3N_4 (SN) used in this study was in house prepared. The size of the SN blocks was 20×20×8 mm and the bend strength was about 700 MPa. The faying face of the blocks was roughly polished with 280 grit SiC. Glass slurries were prepared by mixing Y_2O_3, Al_2O_3, SiO_2 and Si_3N_4 powders thoroughly in a silicon nitride mortar. The glass solders were fabricated in situ on the surface of the ceramic to be

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PII: S0272-8842(98)00007-8

joined during joining experiment. The glass compositions and their abbreviation are listed in Table 1.

2.2. Joining experiments

The test sample for a butt joint, consisting of two blocks of Si_3N_4 separated by a layer of the glass slurry, was fixed in BN powder bed. The samples were heated to a fixed temperature at 10°C min^{-1}, and cooling after holding for a certain time.

2.3. Mechanical tests and microscopy

After joining, specimens were cut perpendicularly to the joint interfaces to prepare bending bars for evaluating the bond strength. The size of the test bars is 3×4×36 mm [3]. The joining strength of the butt joint was determined by a three-point bend test and was calculated as follows

$$\sigma = \frac{3LP}{2t^2 W} \tag{1}$$

where L, t, and W are the sample length, thickness, and width, respectively. P is the load applied when fracture occurs. The standard deviation of the bond strength was then calculated

$$S = \left[\frac{\sum \sigma^2 - \left(\sum \sigma\right)^2/n}{n-1}\right]^{\frac{1}{2}} \tag{2}$$

where n is the number of specimens tested which is five in this paper. Identification of crystals in the glass adhesives was carried out by X-ray diffraction. The microstructures of the joint were investigated by scanning electron microscope (SEM) and optical microscopy.

3. Results and discussion

3.1. XRD result of the glass adhesives

Fig. 1 shows X-ray diffraction patterns obtained from the glass adhesives fired at 1600°C for 30 min then furnace cooled. The heat-treatment is similar to that applied to the joining experiment. In Fig. 1(a) peaks are identified β-$Y_2Si_2O_7$, while in Fig. 1(b) amorphous structure occurs, and in Fig. 1(c) those of $Y_{10}(SiO_4)_6N_2$ (H phase) are identified. Clearly, the amount of α-Si_3N_4 in the glass adhesives plays a key role in the formation of the final phases. The XRD results showed that for adhesive G0, there was no nitrogen atoms in the crystal phase, indicating a pure oxide glass solder, and for adhesive G5 and G10, nitrogen atoms were contained which meant the formation of oxynitride glass. It is considered that the α-Si_3N_4 dissolved in the oxide glass and boosted its nitrogen content, then crystal phase or oxynitride glass were produced. Compared with oxide glasses, oxynitride glasses exhibit higher viscosity and lower thermal expansion coefficient, doubtlessly, these changes might have an effect on the joint strength.

3.2. Effect of glass composition

The effect of the amount of Si_3N_4 in adhesive on the bond strength is shown in Fig. 2. It is clearly seen that the bond strength of the joints decreases as the Si_3N_4 content increases. The liquid phase region is considered to be more narrow with increasing nitrogen concentration in the M–Si–Al–O–N (M = Y, La, Sm, Nd, Yb, Dy, etc.) system [13], consequently, adhesive G0 has much more amount of liquid phase than the other two at the joining temperature of 1600°C. As a consequence,

Table 1
Chemical composition of adhesives (wt%)

	Y_2O_3	Al_2O_3	SiO_2	Si_3N_4
G0	40	20	40	0
G5	40	20	35	5
G10	40	20	30	10

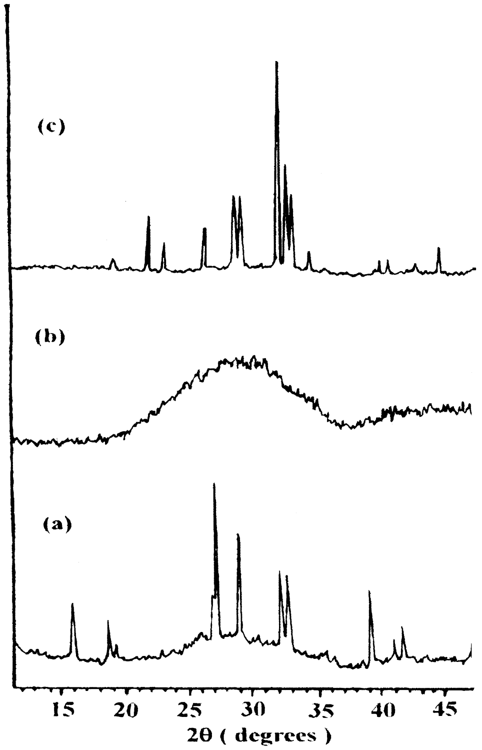

Fig. 1. X-ray diffraction patterns of adhesives GO, G5 and G10. prepared at 1600°C for 30 min, respectively, showing the presence of (a) β-$Y_2Si_2O_7$, (b) amorphous phase, and (c) $Y_{10}(SiO_4)_6N_2$.

adhesive G0 shows lowest viscosity and consequently, best fluidity and wettability, which helps to obtain uniform distribution of the adhesive on the surfaces to be joined and produce an intimate bond. Although the thermal expansion coefficients of G5 and G10 may decrease by the addition of Si_3N_4 powder, matching closely to that of Si_3N_4 ceramic, simultaneously, the amount of solid phase in adhesives G5 and G10 increases, which limits the fluidity and wettability of the adhesives. As a result, the bond strength depends mainly on the wettability and fluidity of the adhesive when joining under pressureless condition.

3.3. *Effect of joining temperature*

In Fig. 3, the measured bond strength for Si_3N_4 joints with adhesive G0 has been plotted versus the joining temperature. The joining was performed at temperatures ranging from 1450 to 1750°C for 30 min. As can be seen, the bond strength of the joint had a maximum value at a joining temperature of 1650°C. At temperatures below 1450°C, a bond could not develop because the adhesive did not melt completely and its viscosity was high. With an increase in temperature, the adhesive dissolved fully, its viscosity reduced and its fluidity improved, moreover, the components of the adhesive diffused rapidly and easily in the liquid. Accordingly, the bond strength markedly increased when the joining temperature increased. As the temperature further increased, the fluid glass drained into the Si_3N_4 and flew out of the contacting region, thus little glass remained between the adherends resulting in a weak bond. In addition, the lower bond strength produced at temperatures exceeding 1650°C might be in part attributed to fact that the magnitude of the residual stress in the joint greatly improved with increasing temperature.

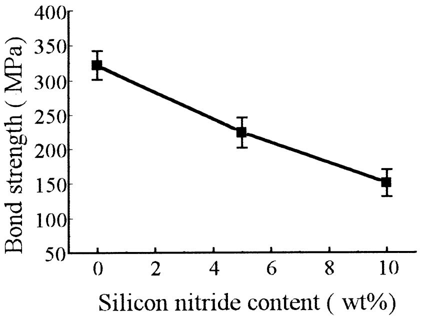

Fig. 2. Effect of silicon nitride content on the bond strength of the joints.

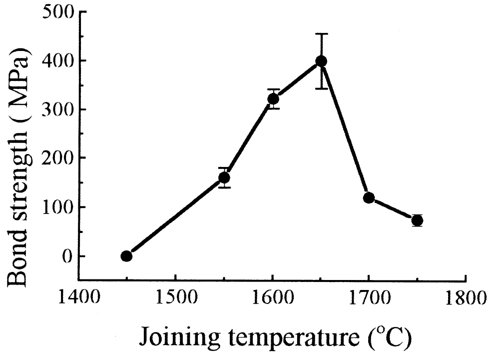

Fig. 3. Dependence of temperature on bond strength.

3.4. *Effect of hold time*

Optical microstructures of samples joined using G0 at 1600°C are given in Fig. 4. The result shows that the thickness of the joints decreases with increasing hold time, indicating densification of the adhesive. Fig. 4 presents the bond strength as a function of hold time, the joining temperature is 1600°C. It is evident that the bond strength improved with increasing hold time for all the three adhesives. For sample joined using G0, its bond strength changes little when the hold time is over 30 min, indicating 30 min at temperature is sufficient to obtain enough bond strength.

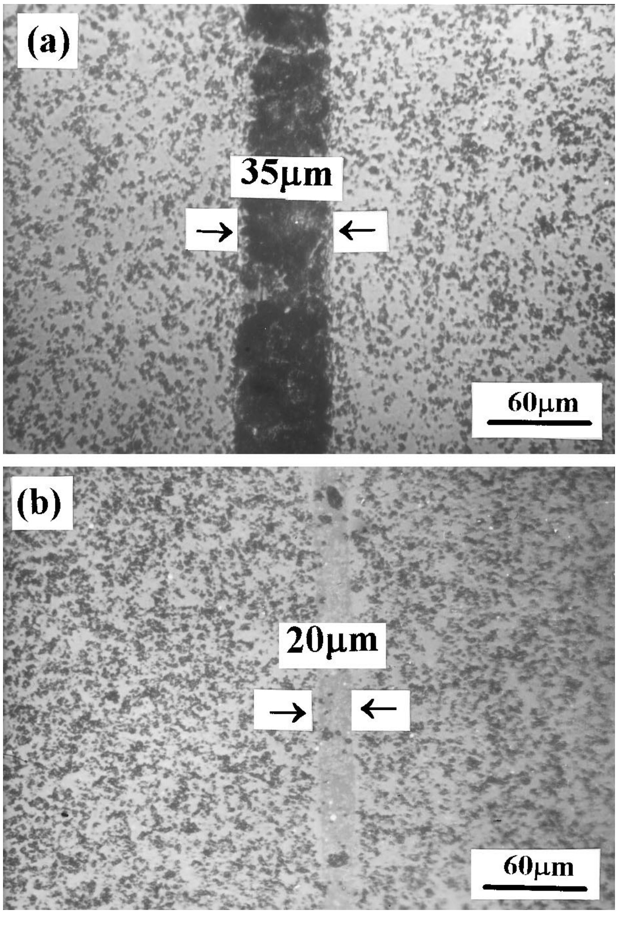

Fig. 4. Optical micrographs of the samples joined at 1600°C for different times, showing the joint thickness of (a) 35 μm and (b) 20 μm.

3.5. Microstructure observations

To get some ideas of the microstructures of the joint, the polished samples were etched in molten NaOH melt for about 2 min. The SEM micrographs of the joint were shown in Fig. 6. Note that the interfacial contact is excellent and the joint is filled with glass. During the joining process, silicon nitride dissolved in the liquid glass, which resulted in silicon nitride grains growing into the joint and interlocking with the glassy bond. The intergrowth network microstructure might be of benefit to the bond strength. However, quite a few thermal cracks in the joint are clearly seen from the pictures. This is caused by the mismatched thermal expansion coefficients between silicon nitride and the glass.

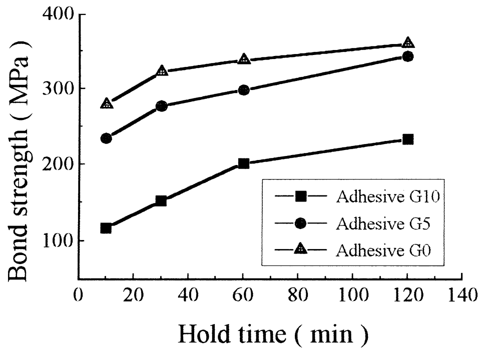

Fig. 5. Effect of hold time on the bond strength of the joints.

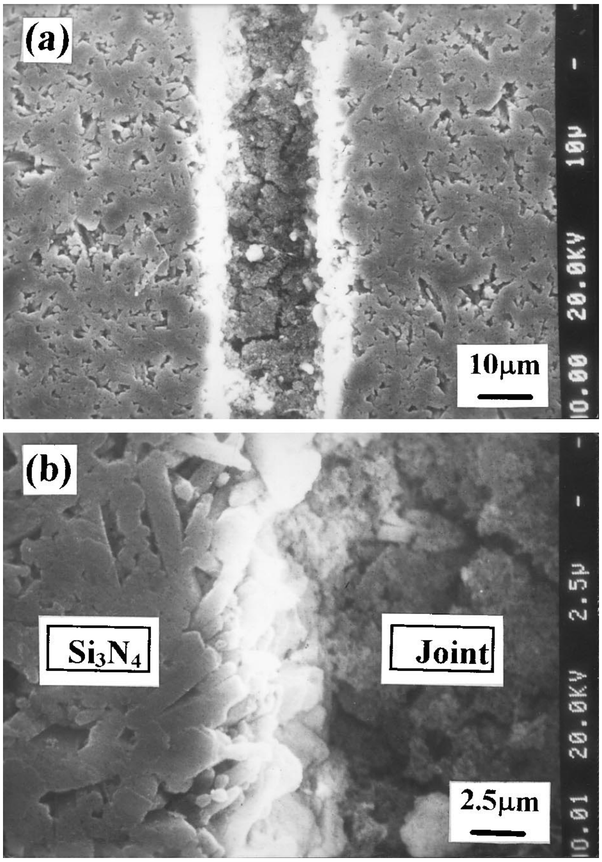

Fig. 6. SEM micrographs of the etched samples prepared at 1600°C for 30 min showing glass-filled joint and thermal cracks.

4. Conclusions

The following conclusions could be drawn on the basis of the above discussions.

1. Under pressurelessly joining conditions, the joint strength decreased sharply with the addition of α-Si_3N_4 to the adhesive due to the high viscosity of the adhesive.
2. An optimum strength of 400 MPa could be obtained when joining was carried out at 165°C for 30 min.
3. An intimate and glass-filled bond was produced using adhesive G0, while many thermal cracks were also observed in the joint.

Acknowledgements

The athors are grateful for the financial support from the Natural Science Foundation of Shanghai under contract no. 97ZE14037.

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