ELSEVIER

Ceramics International 25 (1999) 535–538

CERAMICS
INTERNATIONAL

Bonding silicon nitride using Y_2O_3–Al_2O_3–SiO_2 adhesive

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Received 28 April 1997; accepted 2 September 1997

Abstract

Pressureless sintered silicon nitride ceramics were joined using mixed Y_2O_3, Al_2O_3, SiO_2 and Si_3N_4 powers. Joining was carried out at 1450 and 1600°C for 30 min, under an applied pressure of 5 MPa. The result of EPMA in the cross-section of the joined region indicated that some elements of adhesive (Y, Al) diffused into Si_3N_4 to form a diffusion layer at lower temperature (1450°C), but elements of Y and Al were distributed uniformly all over the cross-section of the joined samples at higher temperature (1600°C). From the results of 3-point bending measurement, an average strength of 400 MPa was obtained for the samples bonded at 1600°C, which was approximately 57% of that of unbonded ceramics.

1. Introduction

Due to their strength, toughness, creep and oxidation resistance and thermal shock properties, silicon nitride ceramics are of great interest for high temperature structural application in advanced heat engines. However, the fabrication of reliable components of silicon nitride with fairly large size and complex geometry for heat engine applications is not possible with today's technology such as injection moulding and pressure casting. Consequently, joining of simpler parts is an attractive alternative.

At present, brazing and diffusion bonding are two principal methods used to bond silicon nitride ceramics [1]. They have common in that the two methods usually use metals or alloys as the interlayer materials, and form an intimate bond by chemical reaction between the interlayer materials and silicon nitride or by the diffusion of metals or alloys into the ceramics to be joined. The great difference of thermal expansion coefficient between the metals and silicon nitride can cause thermal stress in the joint zone, leading to the deterioration of the bond strength. On the other hand, due to the low melting temperature of the interlayer materials, the strength of the joined ceramics begins to decrease sharply at 600°C [2], which limits their high temperature applications. So, it is necessary to develop new joining adhesives so as to minimise the shortcomings mentioned above.

A method of Si_3N_4 joining with oxide glasses was developed [3–9], and the joining mechanism was described elsewhere [9,10]. However, ceramic bonded with pure oxide glass is more likely to break at the joint, because the layer of glass between the adherends has low resistance to crack propagation. Compared to the oxide glasses, the oxynitride glasses have superior mechanical properties such as great hardness and mechanical strength, and better resistance to thermal and mechanical shock [2,11]. Especially, the thermal coefficient of expansion (TCE) of oxynitride glasses decreases as increasing nitrogen content in them, which leads to match closely to that of silicon nitride ceramics. We have succeeded in getting strong bond strength of Si_3N_4–Si_3N_4 ceramic joints at room temperature by using Y–Si–Al–O–N oxynitride glass adhesive with nearly eutectic composition. Therefore, the results obtained were described in this paper.

2. Experimental procedure

2.1. Materials

A sintered silicon nitride (Shanghai Institute of Ceramics) using Y_2O_3 and Al_2O_3 as sintering aids was chosen as the adherend materials. Each material was cut into plates approximately $20\times20\times$ 8 mm then ground by using #600 diamond wheels until flat surface was obtained.

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PII: S0272-8842(97)00091-6

Table 1
Chemical composition of the solder investigated

	Y_2O_3	SiO_2	Al_2O_3	Si_3N_4
(Wt%)	41.5	28	26.5	40

The adhesive material with nearly eutectic composition was prepared from Si_3N_4, SiO_2, Al_2O_3 and Y_2O_3 powders using ball-milling techniques. The chemical composition of the solder investigated is shown in Table 1.

2.2. Joining experiment

Samples are cleaned in water-free ethanol and a slurry of the powdered adhesive is applied to the 20×8 mm surface of the ceramic materials. Silicon nitride ceramics to be joined were packed with BN power in a graphite furnace. The joining was performed at 1450 and 1600°C for 30 min in 0.1 MPa N_2 atmosphere. The heating rate was $10°C\,min^{-1}$. A constant pressure of 5 MPa was applied to aid the joining process.

2.3. Mechanical tests

Joined bars were surface ground and cut into bend test bars $\approx$ 36 by 3 by 4 mm. The bending strength of the joined samples was measured by 3-point bending machine Instron 1195 with span length of 30 mm at a displacement rate of $0.5\,mm\,min^{-1}$.

2.4. Microscopy

The joint region in both broken and unbroken bars was examined by means of optical microscope, EPMA and scanning electron microscope (SEM).

3. Results and discussion

3.1. Joint layer

Fig. 1 shows the optical micrographs of the samples which were joined at 1450 and 1600°C for 30 min, respectively. From the pictures, it indicates the great effect of temperature on the joint thickness. At 1450°C, the joint thickness is about 80 μm, and the joint region consists of diffusion layer and joined layer. While at higher temperature, only a joined layer with the thickness of about a 3 μm exists.

Quantitative analyses elements Y and Al in Si_3N_4/Si_3N_4 joints are shown in Fig. 2. At low temperature that is 1450°C, the yttrium and aluminum contents are higher in the joint compared to the parent silicon nitride ceramics due to the higher proportion of glass found in the joint zone. As shown in Figs. 2(a) and 1(a), Y and Al could penetrate through the grain boundaries into the silicon nitride ceramic to a depth of 60 μm, and their contents are up to the maximum in the joined layer, then decrease along the diffusion layer. However, when the joining temperature of 1600°C is reached, there is no obvious diffusion layer in the joined sample. The distribution of Y, Al and Si elements is uniform all over the cross-section of the joined samples.

In the joining experiment, due to the variation in composition between the grain boundary phase and the joining composition which are both liquids during joining process, there exists a concentration gradient. The chemical potential difference becomes the driving force for diffusion, so elements Y and Al diffuse from the higher concentration zone to the lower one as shown in Fig. 2(a). As the joining temperature increases, the diffusion rate is rapidly improved, simultaneously, it also leads to the decrease of the amount of liquid phase. Finally, the joint zone is only consisted of a very thin joined layer as seen in Fig. 1(b), it means that the

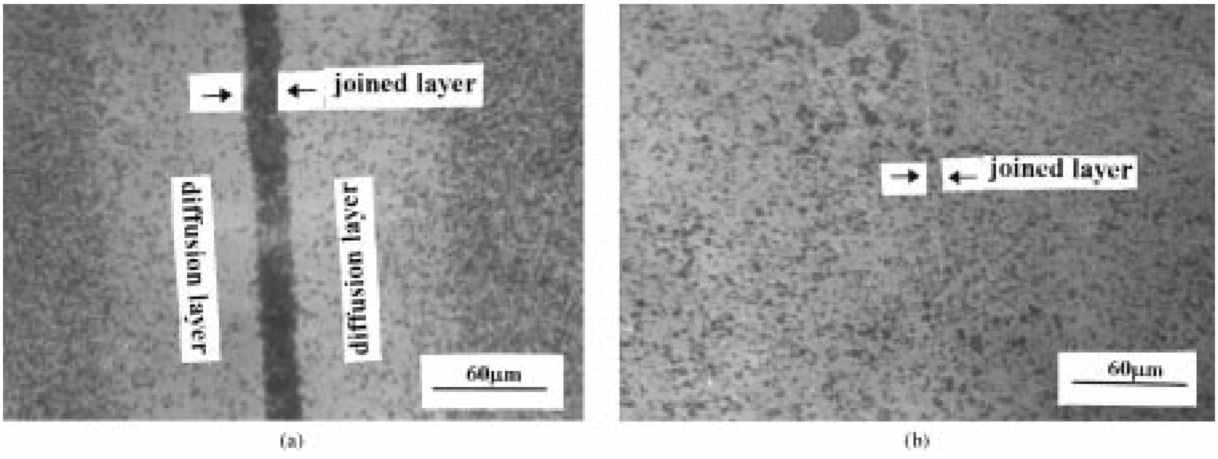
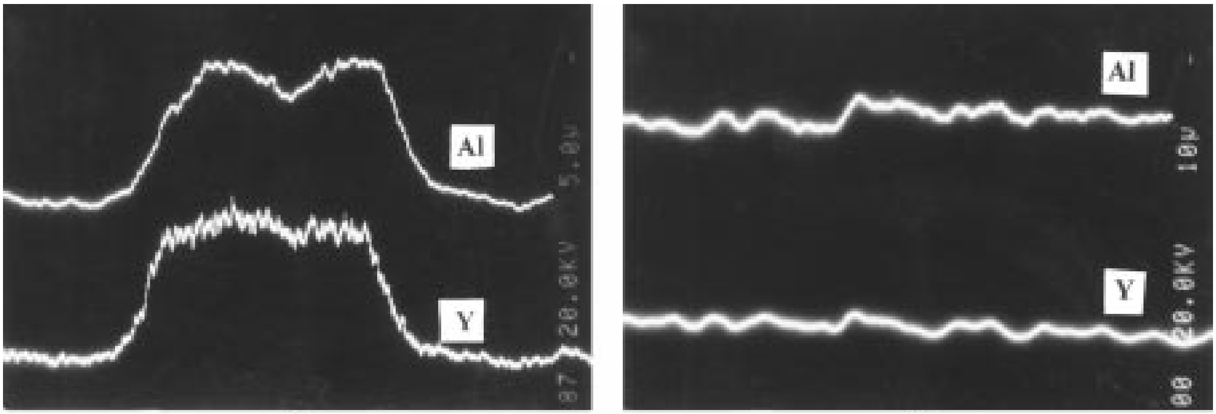


Fig. 1. Optical micrograph of the joined samples: (a) 1450°C; (b) 1600°C.



(a) (b)

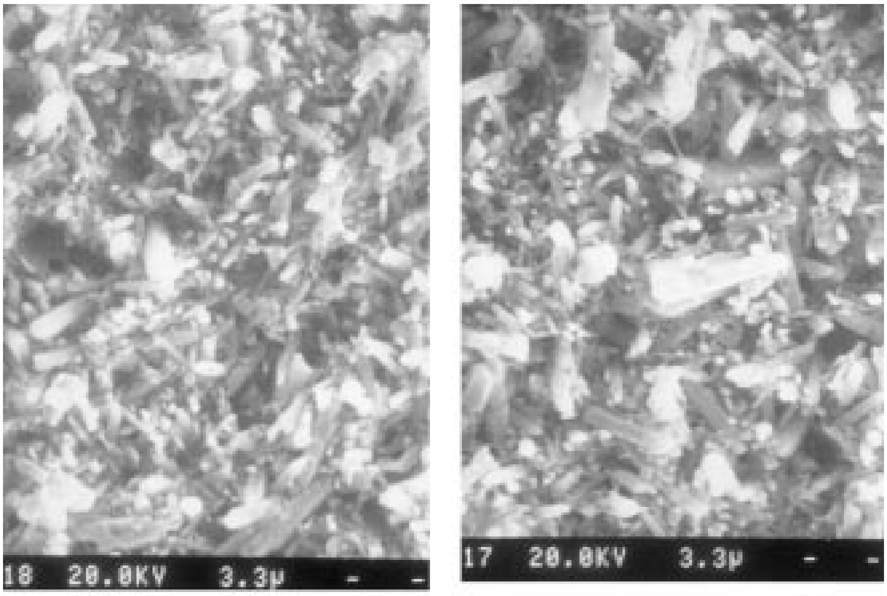
Fig. 2. Line analysis of Y, Al elements around the joined part: (a) 1450°C; (b) 1600°C.

densification of the joining adhesive is nearly completed, which is expected to have better bond strength.

3.2. Bond strength of the joint

The bond strength of the joined samples was evaluated by 3-point bending test. The average and maximum strengths are 400 and 467 MPa, which are 57 and 66% of that of monolithic Si_3N_4 respectively. Fig. 3 shows the microstructures of the joined Si_3N_4 etched by molten NaOH. From the microscopy, it is found that both the joint zone and ceramic matrix have dense microstructure and mainly consist of elongated, needle-shaped β grains. However, the grain size in the joint zone is smaller than that in the ceramic matrix due to the shorter time and lower temperature compared to the ceramic processing.

When the joining temperature is over the eutectic temperature of Y–Si–Al–O–N system, the oxides in the adhesive begin to melt and react to form Y–Si–Al–O liquid phase, which leads to the sintering and densification of the joining solder [12]. As the temperature further increases to 1600°C, α-Si_3N_4 in the adhesive dissolves in the molten glass, boosting the silicon and nitrogen content and altering its composition to sialon glass. Consequently, new β-Si_3N_4 grains begin to precipitation and grow within the joint. Further, acicular β-Si_3N_4 grains form an interlocking network across the joint, at the same time, there are some rearrangement Of β-Si_3N_4 grains at the joint boundary. These lead to the



(a) (b)

Fig. 3. SEM micrograph of the silicon nitride: (a) joined layer; (b) ceramic matrix.

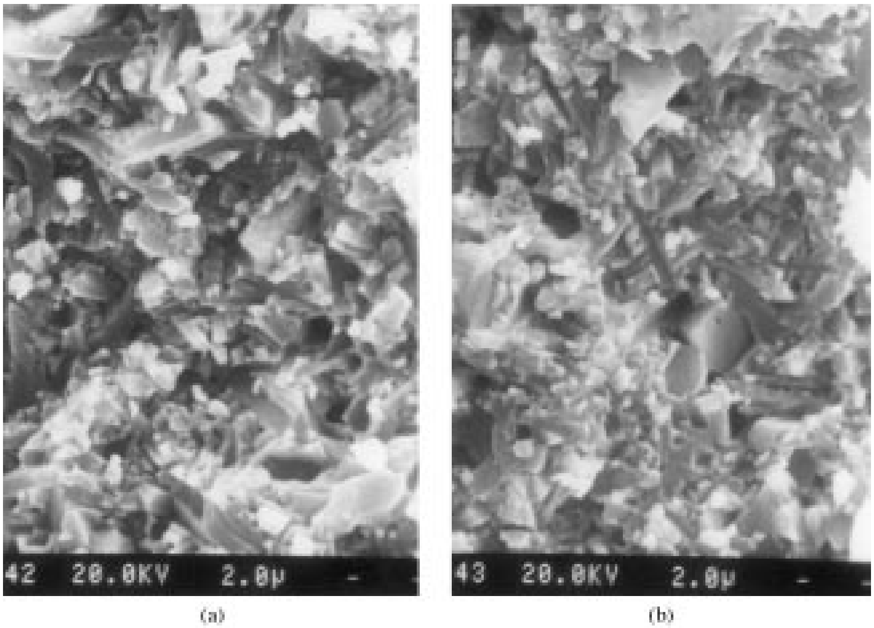


Fig. 4. Fracture surface of the joined samples, showing (a) fracture through Si_3N_4 and (b) fracture through joined layer.

similarity in the microstructure between the joined layer and the original ceramic material. The rearrangement of grains and the intergrowth of the β-Si_3N_4 formed in the adhesive with that in the adherend material aid in strengthening the joint.

3.3. Fracture micrograph

From the results of the 3-point bending test measurement, we learn that the crack initiates in the joined layer, then expands through it; further, it deflects away from the joint and propagates in the Si_3N_4, finally breaks in it. The microstructure of the fracture face is shown in Fig. 4. It can be seen that the microstructure of the joint zone is the same as that of the ceramic matrix, this result has been obtained from the analysis of Fig. 3, and no void and crack are observed in the joint zone.

4. Conclusions

1. Oxynitride glass forming adhesive was successfully used to bond pressureless sintered silicon nitride ceramics.
2. The microstructure of such joint consisted of a very thin layer with a thickness of about 2 μm. The similarity in the microstructure between the joined layer and the bulk ceramic resulted in the strong bond strength.
3. Under the joining condition, the maximum and average strengths of 467 and 400 MPa, i.e., 66 and 57% of that of unbonded materials in the as-sintered state, were obtained.

Acknowledgements

This work was financially supported by Natural Science Foundation of Shanghai under contract no. 97 ZE 14037 in 1997.

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