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Diffusion bonding of ceramics: mullite, ZrO_2-toughened mullite

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Diffusion bonding of fine-grained mullite and ZrO_2-toughened mullite was performed in the temperature range from 1500 to 1550 °C in air. Uniaxial pressure was applied at high temperature during the bonding process. The surface roughness to be bonded (R_{max}) was about 3 μm. Bonding strength was measured by four-point bending tests and the strength of the base material was measured by three-point bending tests. The effects on the bonding strength of bonding conditions such as temperature and applied strain were examined. Bonding strength increased with increasing bonding temperature and applied strain. The bonding strength of mullite and ZrO_2-toughened mullite was about 80% of the strength of the base material before bonding. The bonding strength of mullite was maintained up to 1000 °C.

1. Introduction

Bonding methods for ceramic–ceramic joints [1–4] and ceramic–metal joints [5, 6] have been developed to fabricate large, complex-shaped components. Diffusion bonding is a solid-state bonding method which is advantageous because the strength of the bonding zone can reach a value which is almost equal to that of the base material. The process of diffusion bonding requires close physical contact at the interface by (i) plastic deformation (power-low creep and/or superplastic flow) of surface asperities, and (ii) void elimination through diffusion [7–9].

Diffusion bonding of superplastic titanium alloy [9] has attracted industrial interest because complex components are produced by superplastic forming concurrent with diffusion bonding (SPF/DB process). The authors [10–12] recently performed diffusion bonding of superplastic ceramics (Y_2O_3-stabilized tetragonal ZrO_2 polycrystals [13], ZrO_2–Al_2O_3 composites [14, 15] and ductile Al_2O_3 [16]). The bonding strength for similar materials was 1300 MPa, and that for dissimilar materials was 1200 MPa. Since the microstructural requirement for superplasticity is a very fine grain size, it was supposed that the diffusion bonding of fine-grained ceramics would yield good results.

Recent advances in powder processing made it possible to synthesize stoichiometric ultra-fine powders of mullite ($3Al_2O_3 \cdot 2SiO_2$) by the metal alkoxide process [17], spray pyrolysis [18], and the sol–gel method [19]. High-purity mullite is recognized as a promising structural ceramic due to its mechanical properties at elevated temperatures [18, 20]. In this paper we studied diffusion bonding of fine-grained mullite and ZrO_2-toughened mullite [21] which were prepared from mullite powder [19] of stoichiometric composition by the sol–gel method.

2. Experimental procedure

2.1. Materials

The materials used in bonding tests were stoichiometric mullite and ZrO_2-toughened mullite (MP-20 and MZ-15, respectively, from Chichibu Cement Co., Kumagaya, Japan) in which 15 vol % ZrO_2 grains were dispersed among mullite grains. The sol–gel-derived mullite powder [19] was sintered at 1610 °C in air for 3 h. The mullite–ZrO_2 gel powder was sintered at 1650 °C in air for 2 h [21]. Some properties of the materials are summarized in Table I. The ZrO_2-toughened mullite exhibited superplastic elongation of 120% in preliminary tension tests at 1550 °C and $2.86 \times 10^{-5} s^{-1}$ [22]. The as-obtained ceramic bodies were ground with a No. 200 diamond wheel. The surfaces to be joined were finished with a No. 400 diamond wheel. The surface roughness (R_{max}) was about 3 μm. The size of the specimens for bonding tests was 15 mm × 25 mm × 20 mm.

2.2. Bonding procedure

The bonding between similar materials was performed by using a universal testing machine in air. Diffusion bonding of ZrO_2-toughened mullite was conducted at

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TABLE I Properties of materials

Material	Composition (wt %)						
	Al_2O_3	SiO_2	ZrO_2	Density ($g\,cm^{-3}$)	Grain size (μm)	Bending strength (MPa)	Thermal expansion ($10^{-6}\,°C^{-1}$)
Mullite	71.8	28.2	–	3.11	1.79	280	4.5
Mullite–ZrO_2	53.7	21.1	25.2	3.55	0.81	410	4.9

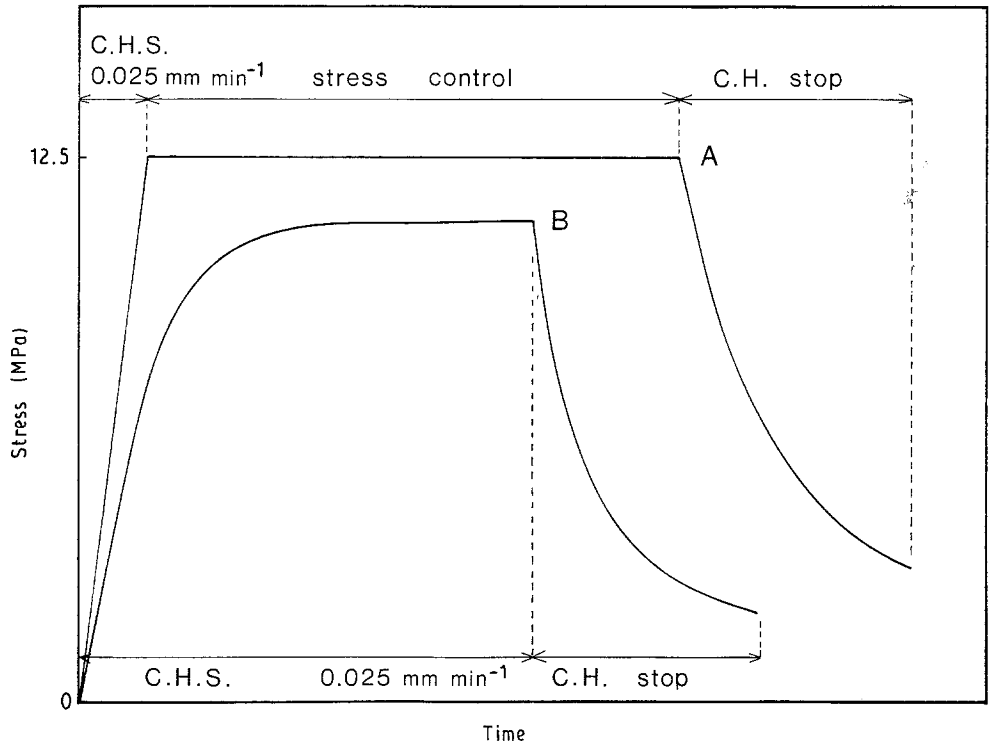

Figure 1 Stress–time curves in bonding procedure (C.H.S. = crosshead speed).

1500, 1525 and 1550 °C, while diffusion bonding of mullite was conducted at 1550 °C only. The bonding procedure is illustrated in Fig. 1. Two specimens were heated and pressed together in an SiC jig at a crosshead speed of 0.025 mm min^{-1} up to a compressive stress of 12.5 MPa. A constant stress was then held by controlling the movement of the crosshead. When the deformation of the specimen reached a certain strain, the crosshead was stopped and held for 30 min (line A in Fig. 1). When plastic deformation occurred at stresses lower than 12.5 MPa, the specimens were deformed at constant crosshead speed to a certain strain. The crosshead was then stopped and held for 30 min (line B in Fig. 1).

2.3. Evaluation of bonding strength

The bonded materials were cut into rectangular bars 3 mm × 4 mm × 40 mm by a diamond cutter; the specimens were then ground with Nos 200 and 400 diamond wheels. The bonding strength was measured at room temperature by a four-point bending test (lower span = 30 mm, upper span = 10 mm) at a crosshead speed of 0.5 mm min^{-1}.

2.4. Evaluation of strength of base material

The strength of the base material was evaluated by a three-point bending test (span = 16 mm, crosshead speed = 0.5 mm min^{-1}) by using specimens which had been broken into two pieces in the bonding strength test.

3. Results and discussion

3.1. Bonding

A typical stress–time curve in bonding tests was line A as shown in Fig. 1. The stress–time curve of line B was observed only when the ZrO_2-toughened mullite was bonded at 1550 °C. The materials were bonded with each other under all the conditions in these experiments.

3.2. Microstructure at bonding interface

Scanning electron micrographs of bonding interface between mullite parts are shown in Fig. 2. Voids at the bonding interface decreased with increasing applied strain. As small voids still remained at the interface of

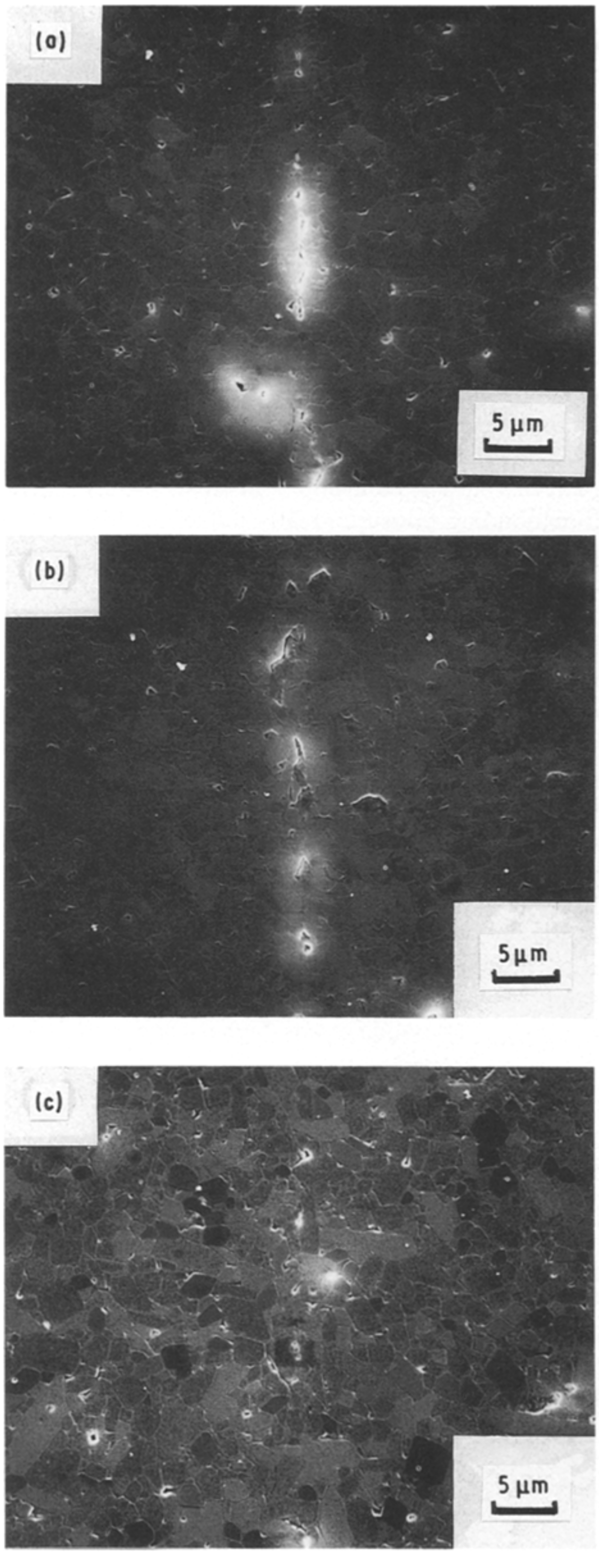

Figure 2 Scanning electron micrographs of interface between mullite parts bonded at 1550 °C: strain (a) 2.5%, (b) 5%, (c) 10%.

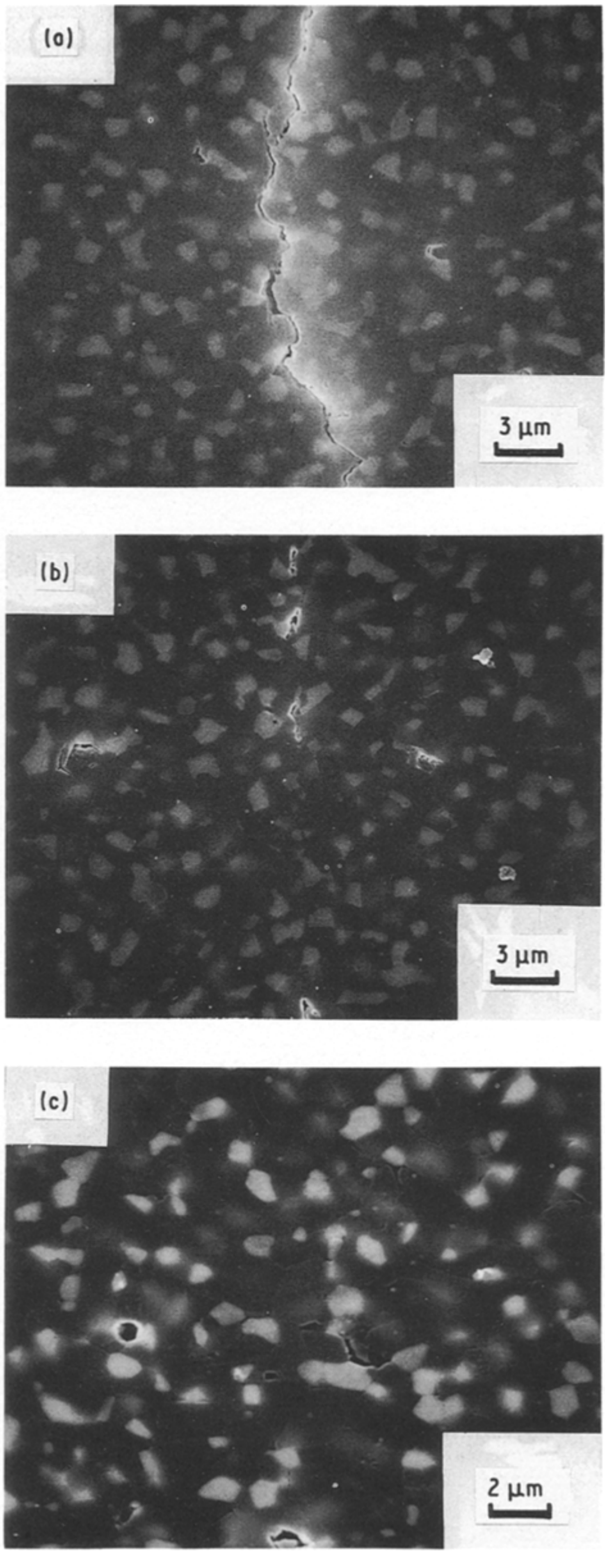

Figure 3 Scanning electron micrographs of interface between ZrO_2-toughened mullite parts: (a) 2.5% strain at 1500 °C, (b) 2.5% strain at 1550 °C, (c) 10% strain at 1550 °C.

specimens which were deformed by 10%, the bonding must be performed at higher temperature, higher pressure or higher strain to obtain complete bonding. Scanning electron micrographs of the bonding interface between ZrO_2-toughened mullite specimens are shown in Fig. 3. The bonding process at 1550 °C (Fig. 3b) proceeded faster than that at 1500 °C (Fig. 3a), because the plastic deformation resistance at 1500 °C was higher than that at 1550 °C and the contact process of bonding did not progress smoothly. The voids at the bonding interface decreased (Fig. 3b and c) with increased applied strain.

3.3. Bonding strength

The relationship between bonding strength and applied strain of mullite which was bonded at 1550 °C is shown in Fig. 4. The bonding strength increased with strain. The bonding strength of mullite at high temperature is shown in Fig. 5. All bonding specimens were broken at the bonding interface in four-point bending tests. The bonding strength was maintained up to 1000 °C and decreased to 120 MPa at 1400 °C.

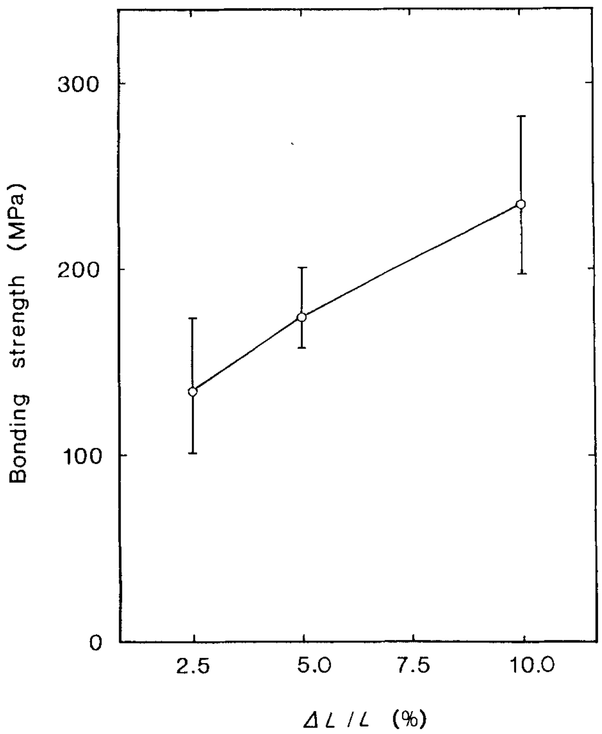

Figure 4 Relationship between bonding strength and applied strain of mullite bonded at 1550 °C.

The effects of applied strain and bonding temperature on bonding strength is shown for ZrO_2-toughened mullite in Fig. 6. Generally the bonding strength increased with increasing bonding temperature. When the bonding temperature was 1550 °C (strains of 2.5 and 5%), however, the bonding strength was lower than the value obtained at 1525 °C. ZrO_2-toughened mullite was deformed superplastically at 1550 °C at a lower stress than 12.5 MPa (line B in Fig. 1). The bonding was also conducted at higher crosshead speed (0.05 mm min^{-1}) to increase the applied stress. The results are shown in Fig. 7. There was not much difference on changing the crosshead speed at large strain. On the other hand, the bonding strength at a crosshead speed of 0.05 mm min^{-1} was higher than that at 0.025 mm min^{-1} at small strains. Therefore, bonding pressure greatly influences the contact process of bonding.

3.4. Strength of base material

The relationship between the strength of the base material after bonding and applied strain is shown for mullite in Fig. 8. The bending strength of the base material after bonding was higher than that before bonding, but was not dependent significantly on the difference in applied strain. The effects of applied strain and bonding temperature on the strength of the base material are shown for ZrO_2-toughened mullite in Fig. 9. The strength of the base material increased with bonding temperature when compared at the same strain. In particular the strength of the base material after bonding was higher than that before

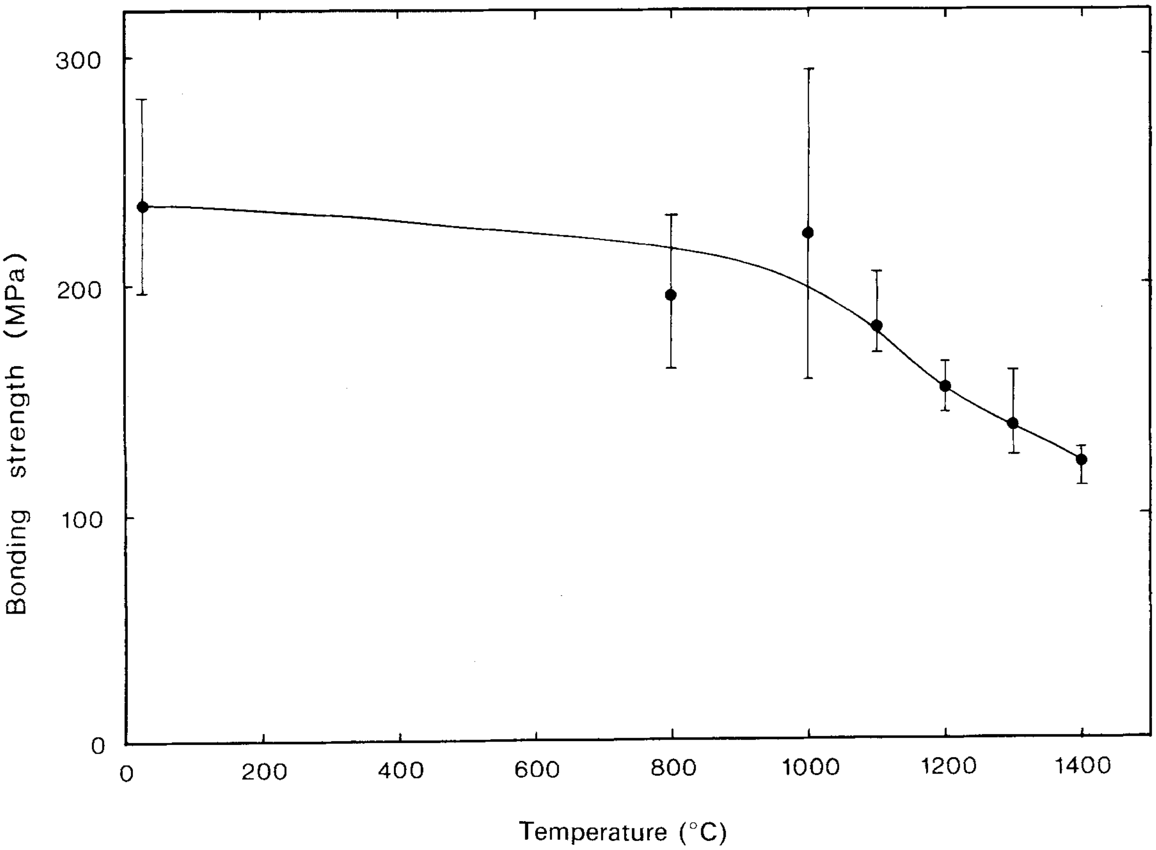

Figure 5 Bonding strength of mullite at high temperature.

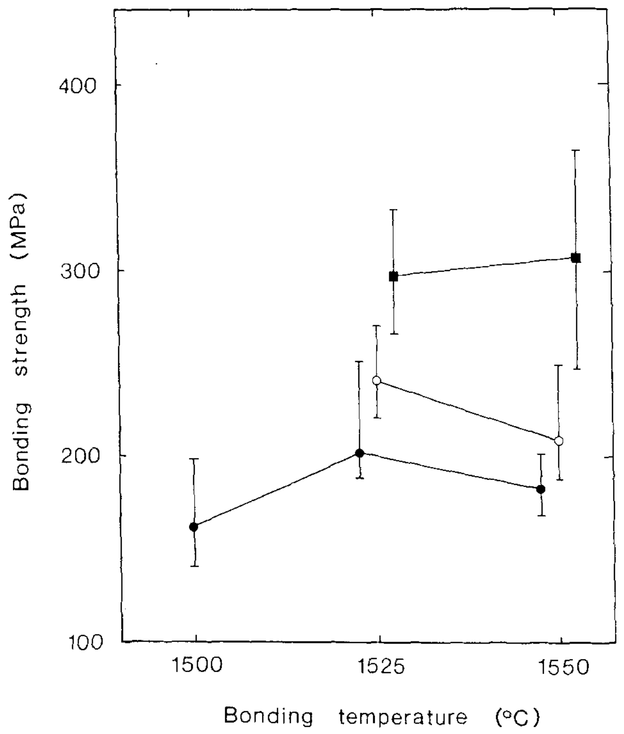

Figure 6 Effects of applied strain and bonding temperature on bonding strength of ZrO_2-toughened mullite: $\Delta L/L$ (%) = (●) 2.5, (○) 5.0, (■) 10.0.

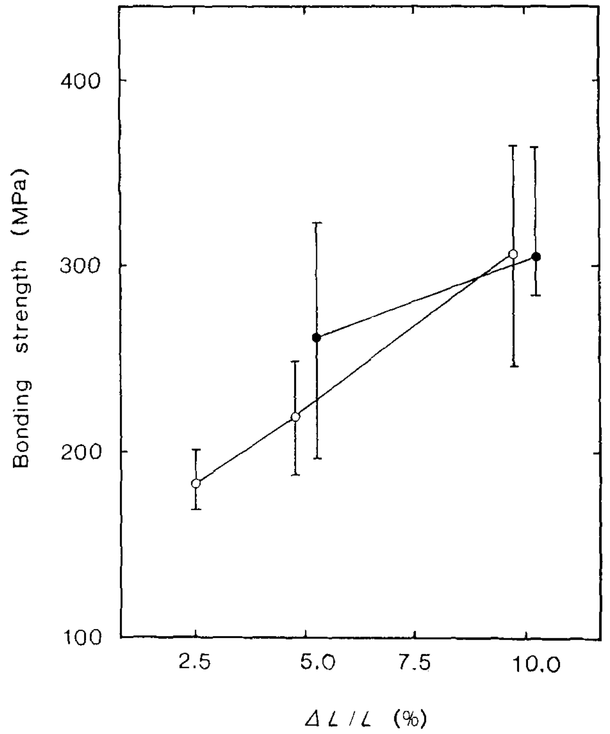

Figure 7 Dependence of strain on bonding strength of ZrO_2-toughened mullite at crosshead speed of (○) 0.025 and (●) 0.050 mm min^{-1}.

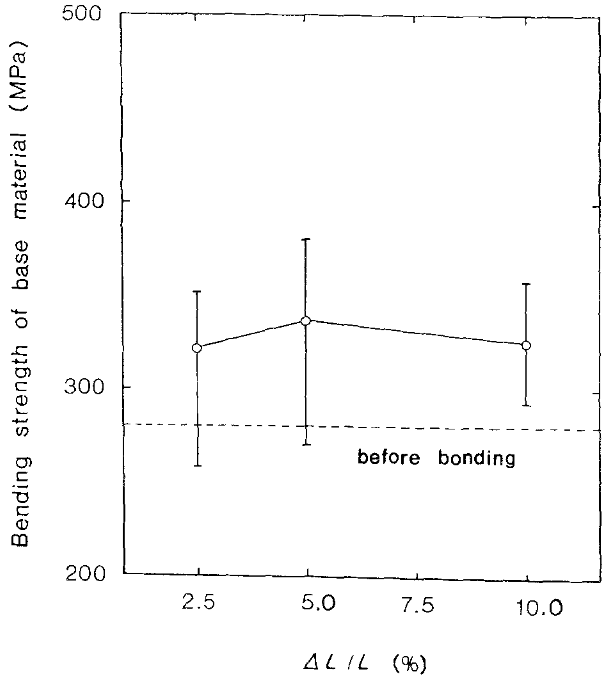

Figure 8 Relationship between the strength of the base material after bending and applied strain of mullite.

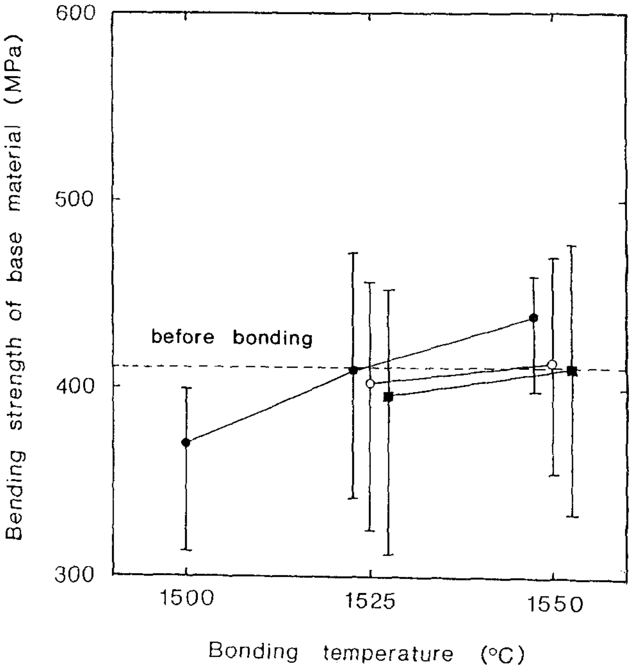

Figure 9 Effects of applied strain and bonding temperature on the strength of base material of ZrO_2-toughened mullite: $\Delta L/L$ (%) = (●) 2.5, (○) 5.0, (■) 10.0.

bonding for a strain of 2.5% at 1550 °C. The strength of the base material decreased with increasing strain at every bonding temperature. Measurements of density and grain size and X-ray diffraction analysis on all specimens were performed to study the influence of compression at high temperature on the microstructure of the base material. No differences between specimens before and after bonding were detected by any measurement. The authors believe that the strength of the base material after bonding increased due to the reduction of defect size by compression at high temperature.

4. Conclusion

1. Bonding strength was affected by bonding temperature, strain, bonding pressure and crosshead speed.

2. The strength of the base material was affected by bonding temperature, strain and bonding pressure.

The strength of mullite base material after bonding was higher than that before bonding under all bonding conditions. The strength of ZrO_2-toughened mullite base material after bonding was higher than that before bonding when bonding was conducted by applying a strain of 2.5% at 1550 °C.

3. The bonding strength of mullite was 240 MPa when bonding was conducted by applying a strain of 10% at 1550 °C, and that of ZrO_2-toughened mullite was 310 MPa.

4. The bonding strength of mullite was maintained up to 1000 °C.

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