JOURNAL OF MATERIALS SCIENCE 29 (1994) 5041–5046

The influence of brazing conditions on joint strength in Al_2O_3/Al_2O_3 bonding

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The brazing of alumina ceramic to itself was performed using $Ag_{57}Cu_{38}Ti_5$ filler alloy. The bonding was carried out in a vacuum of 7×10^{-3} Pa, and the joining conditions were at 1073, 1123, 1173, 1223, 1273 and 1323 K for 1.8 ks under a pressure of 0.01 MPa, at 1123 K with a pressure of 0.01 MPa for 0, 0.3, 0.9, 1.8, 2.7 and 3.6 ks, and at 1123 K for 1.8 ks with pressures of 0, 0.01, 0.05, 0.10, 0.15, 0.20 and 0.30 MPa, to determine the effects of joining temperature, pressure and holding time on the joint strength. The joint strength was measured by shear tests. The interface microstructures and fractured surfaces after testing were observed by scanning electron microscopy (SEM). It was shown that the shear strength of Al_2O_3/Al_2O_3 joints was largely affected by the joining conditions; it first increased and then decreased with increasing joining temperature, pressure and holding time and depended mainly on the strength of interfacial reaction layer itself and the interface bonding strength between the reaction layer and the ceramic. The maximum joint strength was obtained when the reaction occurred under a suitable temperature, pressure and time, and the reaction layer thickness was about 2 μm. SEM observations revealed that there were four types of fracture and each kind corresponded to a different strength.

1. Introduction

Alumina ceramics not only have excellent strength, high resistance to wear and corrosion at elevated temperature, but also have unique properties of electronic insulation, and they are becoming increasingly important in engineering applications as both structural and insulating materials. However, difficulty in the fabrication process of complex-shaped and large-sized components requires techniques of joining ceramics to be developed.

Brazing methods have been widely used in joining ceramics. During the process of brazing ceramics, apart from the properties of joining materials, the joint properties are also sensitive to the brazing procedures [1–6]. The brazing atmosphere, heating and cooling rate, joining temperature, holding time and joining pressure, etc., have a considerable influence on the joint strength, and thus precise process control is required to obtain reliable joints.

On the other hand, most active filler alloys contain titanium. Experimental studies on brazing of non-oxide ceramics have shown that the interfacial reaction between the ceramics and the filler metals plays an important role in determining the joint strength of the brazed interface [6–10]. In our previous work, it was found that the ZrO_2 ceramics can react with Ag–Cu–Ti filler metal and the joint strength is mainly controlled by the interfacial reaction layer thickness and morphology [11]. However, there is some disagreement concerning the interfacial reaction between alumina and titanium-containing filler metal, and its effects on the joint strength are not clear.

In the present study, alumina ceramic was brazed to itself using Ag–Cu–Ti filler metal with varied joining conditions such as joining temperature, pressure and holding time. The relationships of the joining strength and the joining conditions as well as the interfacial reaction, were also analysed.

2. Experimental procedure

The alumina ceramic of 99% purity was used. The filler metal selected was $Ag_{57}Cu_{38}Ti_5$ and its thickness was about 50 μm.

Before joining, the surfaces to be brazed of the ceramics and the filler metal were polished mechanically and then cleaned carefully in acetone. Two pieces of Al_2O_3 discs, 15 and 5 mm diameter and 5 mm thick, were used to make a lap joint using Ag–Cu–Ti filler of 5 mm diameter and 50 μm thick, as shown in Fig. 1a. There were three sets of brazing processes to determine the effects of brazing conditions such as joining temperature, pressure and holding time on the joint strength, as shown in Table I.

The brazing was carried out in a vacuum of about 7×10^{-3} Pa. The heating rate, up to the joining temperature, was 15 K min^{-1}. After bonding, the specimen was cooled slowly to room temperature at a rate of 3–5 K min^{-1} to 873 K and then furnace cooled to room temperature. The strength of the joints was

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measured by shear tests using compressive loading at room temperature at a crosshead speed of $0.5\ \mathrm{mm\,min^{-1}}$ with the apparatus manufactured as shown in Fig. 1b. The shear strength of the lap joints was determined by dividing the fracture load by the cross-sectional area of the joints.

The microstructures and fractured surface of the joints after applying the shear testing were observed and analysed by scanning electron microscopy (SEM). The details of interfacial analyses have been described previously [12].

3. Results and discussion

3.1. Effects of joining temperature

Fig. 2 shows the effect of joining temperature on the joint strength and reaction-layer thickness under a pressure of 0.01 MPa brazing for 1.8 ks (Set 1). It can be seen from the figure that when the joining temperature was lower than 1123 K, the joint strength rapidly increased with increasing joining temperature. However, it decreased gradually when the joining temperature, was higher than 1123 K. The maximum joint strength was obtained at 1123 K. Interfacial reaction layer thickness was increased with increasing joining temperature, and from 1.2 μm at 1073 K increased steeply up to 8.6 μm at 1323 K. It is obvious that sufficient strength was guaranteed only if a reaction layer with a certain thickness (about 2 μm) was formed.

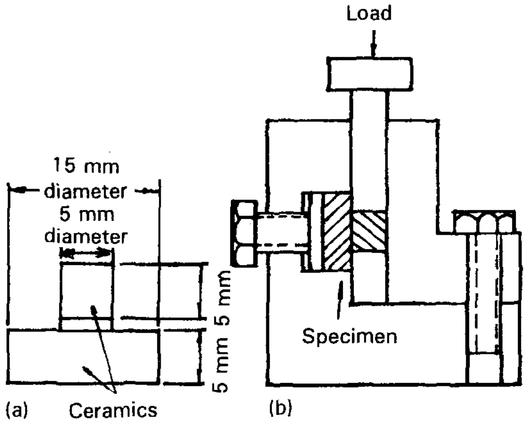

Figure 1 (a) The bonded Al_2O_3/Al_2O_3 assembly, and (b) the test apparatus used for the shear test.

TABLE I Brazing processes

Set	Temperature (K)	Pressure (MPa)	Time (ks)
1	1073–1323	0.01	1.8
2	1123	0.01	0–3.6
3	1123	0–0.3	1.8

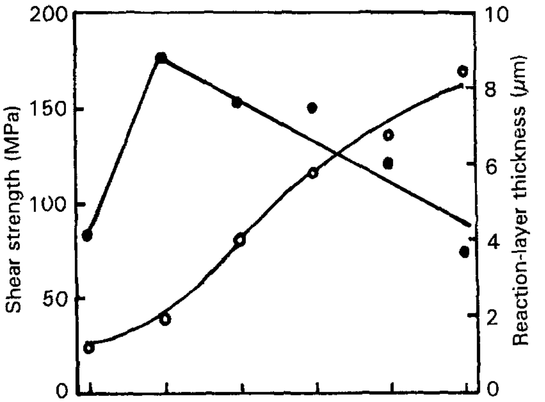

Figure 2 Effect of joining temperature on (●) joint strength and (○) reaction-layer thickness. Holding time 1.8 ks, joining pressure 0.01 MPa.

Scanning electron micrographs of the joining interface are shown in Fig. 3. It is notable that cracks

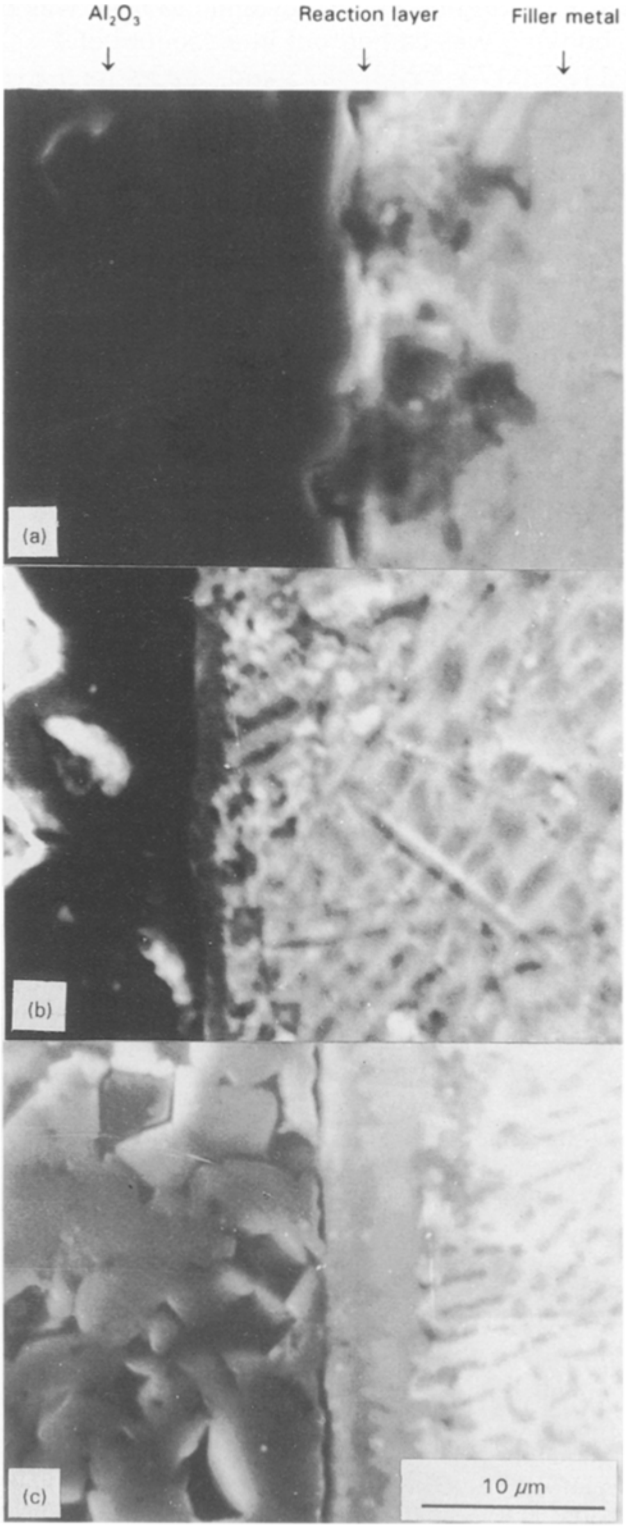

Figure 3 The microstructures of Al_2O_3/Ag–Cu–Ti interface brazing at (a) 1073 K, (b) 1123 K and (c) 1223 K for 1.8 ks.

parallel to the interface were observed at the interface between the reaction layer and the alumina ceramic; these cracks were mainly caused by differences in thermal expansion coefficients between the interface and the ceramic and the large differences of temperature, because of cooling from the joining temperature to room temperature. The real contact area between the alumina ceramic and the reaction layer joined at 1123 K was apparently larger than that joined at 1323 K, thus resulting in a decreased joint strength with the increment of joining temperature.

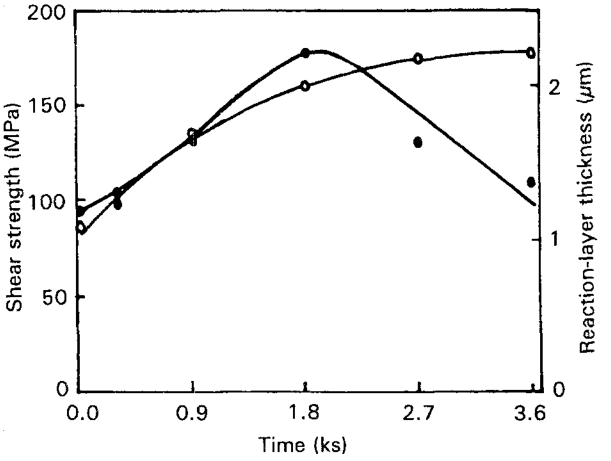

Figure 4 Effect of holding time on (●) joint strength and (○) reaction-layer thickness, brazing at 1123 K with a pressure of 0.01 MPa.

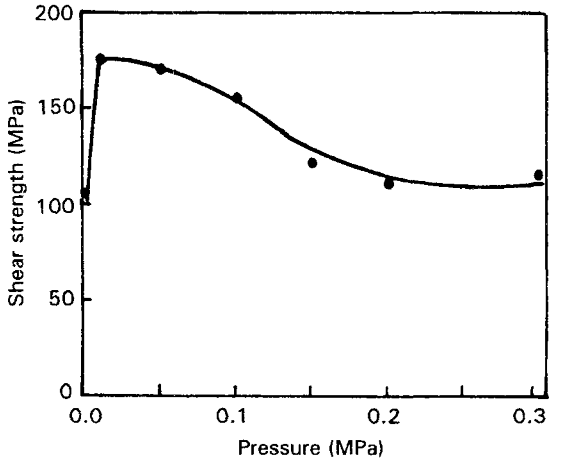

Figure 5 Effect of joining pressure on the joint strength, brazing at 1123 K for 1.8 ks.

3.2. Effect of holding time

Fig. 4 shows the effects of holding time on the joint strength and reaction-layer thickness brazed at 1123 K with a pressure of 0.01 MPa (Set 2). It can be seen that the joint strength first increased and then decreased with increment of holding time, while the reaction-layer thickness was simply increased with increasing holding time, and from 1.0 μm for 0 ks increased up to 2.2 μm for 3.6 ks. The maximum joint strength was achieved when brazing for 1.8 ks and with a reaction layer of 2 μm.

3.3. Effect of joining pressure

The effects of joining pressure on the joint strength (Set 3) is shown in Fig. 5. When the joining pressure was lower than 0.01 MPa, the joint strength increased remarkably, due to the true contact area between ceramic and filler metal being expanded, as shown in Fig. 6. When the joining pressure increased further, it was found that parts of the filler alloy flowed out of the joining interface, and the more the filler metal bled from the interface, the higher the joining pressure became, and thus the filler metals in the interface were decreased and resulted in an insufficient interfacial reaction and hence a decrease in the joint strength.

3.4. Fracture analyses

Typical fracture surfaces of the joints after applying shear testing were observed by SEM. The fracture of these ceramic/ceramic joints can be divided into four types: A, B, C and D, as shown in Fig. 7.

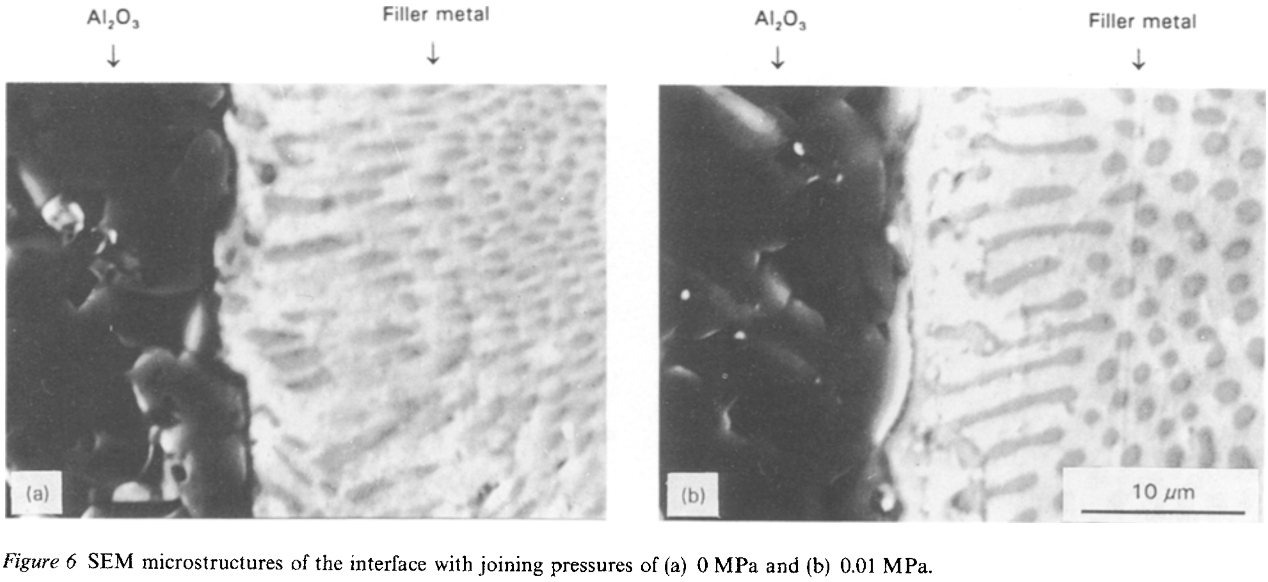

Figure 6 SEM microstructures of the interface with joining pressures of (a) 0 MPa and (b) 0.01 MPa.

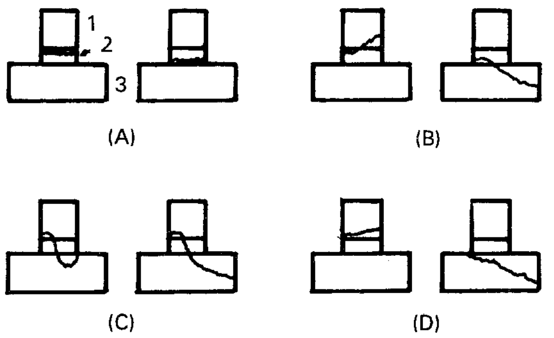

Figure 7 Schematic diagram illustrating the four kinds of ceramic/ceramic joints. 1, 3 ceramic; 2, reaction-layer or brazing filler metal. Fracture occurred (A) at the interface, (B) partially at the interface and partially at the ceramic, (C) a crack started at one ceramic and propagated across the interface into another ceramic, (D) at the ceramic.

Fracture occurred rigorously at the interface, fracture type A. This fracture type corresponded to the low strength of a joint. The poor interfacial bonding strength of the ceramic to the filler metal was mainly due to (i) insufficient interfacial reaction, as at low joining temperature and short brazing time, (ii) interface brittleness because of the presence of more brittle reaction products, such as Ti_2O, TiO, and interface cracks, because of the thermal expansion mismatch at a higher temperature and for a longer holding time.

Cracks started at the interface and propagated into the ceramic, fracture type B. This fracture type corresponded to a medium joint strength of ceramic-to-ceramic joints. SEM fracture surfaces of this kind of fracture are shown in Fig. 8.

Fracture which initiated in a ceramic near to the interface and then propagated across the interface into another ceramic, is termed fracture type C. This fracture type corresponded to a high joint strength. Fig. 9 shows the SEM fracture surface of type C in large ceramic side brazing at 1123 K for 1.8 ks.

Fracture also occurred in ceramics very near to the interface: fracture type D. This fracture type corresponded to a higher joint strength, but the joint strength was still lower than that of the ceramic itself. It is obvious that the strength of the interface between the ceramic and the filler metal was higher than that of the ceramic near the interface, and the ceramic near the interface must have been damaged by residual stress.

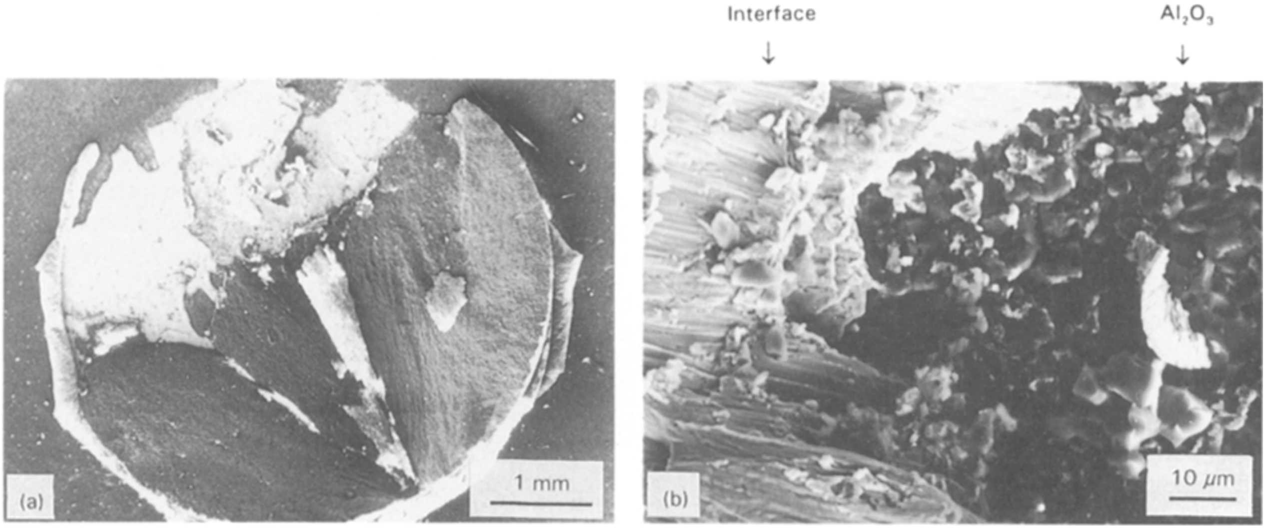

Figure 8 Scanning electron micrographs of the fracture surface of (a) fracture type B and (b) regional magnification of (a) in the large ceramic side, brazing at 1123 K for 0.9 ks.

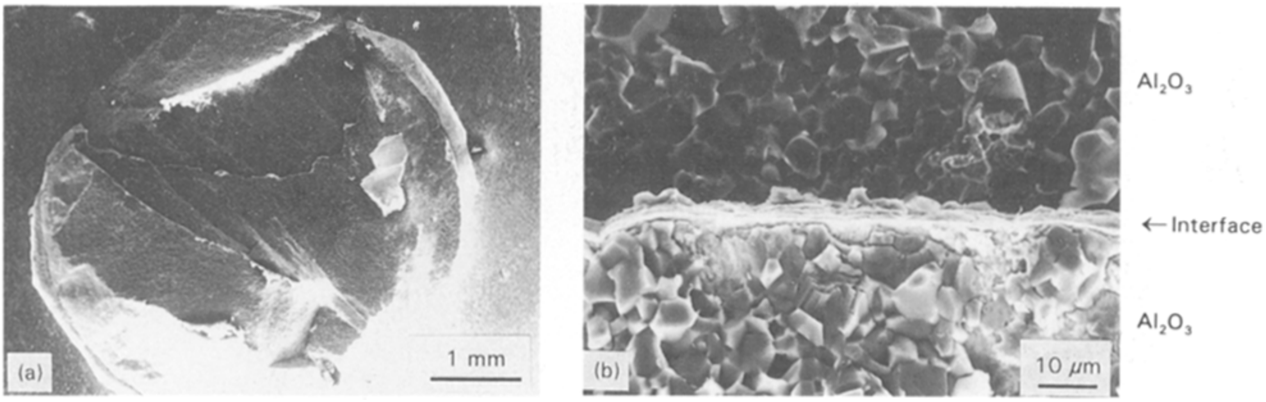

Figure 9 Scanning electron micrographs of the fracture surface of (a) type C, and (b) regional magnification of (a), brazing at 1123 K for 1.8 ks.

3.5. Effect of interface reaction on joint strength

According to X-ray diffraction (XRD) analyses results of reaction products at the interface of Al_2O_3/Ag–Cu–Ti filler metal [12], Al_2O_3 ceramic can react with Ag–Cu–Ti filler metal, producing AlTi and Cu_2Ti_4O at or below 1123 K, and Ti_2O, TiO and $CuTi_2$ above 1123 K, and transition layer structures with $Al_2O_3/Ti_2O + TiO/Ti_2O + TiO + CuTi_2/CuTi_2$/Ag–Cu were formed at the interface. The chemical reaction can be described by the reduction of the alumina with titanium in the filler metal, followed by precipitation of AlTi, Ti_2O or TiO at the interface. Once a continuous AlTi, Ti_2O or TiO thin layer was formed, it was difficult for titanium to diffuse further through the interface, therefore, the titanium concentrated in the vicinity of the AlTi, Ti_2O or TiO layer and reacted with copper or oxygen disolved in the alloy to form Cu_2Ti_4O or $CuTi_2$.

From the above results in Figs 2 and 4, it is revealed that the joint strength first increased and then gradually decreased with increment of joining temperature and holding time, but the reaction layer thickness was simply increased with rising temperature and holding time. The relationship between the joint strength and the reaction-layer thickness is shown in Fig. 10. It can be seen from this figure that the maximum shear strength value was obtained when the reaction-layer thickness was about 2 μm.

Generally, the joint strength at room temperature with a growing reaction layer is thought to be influenced by two factors: interfacial bonding strength and the strength of the reaction layer. Interfacial bonding strength was presumed to be the sum of the chemical bonding strength at the interface and the mechanical interlocking strength of the interface. In the system, the reaction between the ceramic and the filler metal occurred strongly, such as the Al_2O_3/Ag–Cu–Ti joint, and the contribution of chemical bonding to the joint strength is greater than that of mechanical interlocking. The strength of the reaction layer, however, depended mainly on the properties of the reaction products. When the brazing temperature was low and the holding time short, the interfacial reaction between alumina and filler metal was insufficient and both the interfacial bonding and reaction-layer strength were low; therefore, the joint strength was also low. With increasing brazing temperature and holding time, the interfacial reaction became sufficient and both the interfacial bonding and the reaction-layer strength were increased, and the joint strength gradually increased, but when the brazing temperature was too high and the holding time too long, the reaction layer strength was not increased further but decreased, because the amounts of brittle reaction products increased. On the other hand, the true contact area between the reaction layer and alumina ceramic decreased because of the thermal expansion mismatch at the interface and the greater difference between the joining temperature and room temperature, and finally caused a degradation of the joint strength. The strength of the Al_2O_3/Al_2O_3 joint specimens that fractured entirely through the Al_2O_3 were still lower than the average strength of monolithic Al_2O_3. This fact, together with the observation that fractures in the ceramic always occurred near the brazed surfaces indicated that the damage of alumina near the interface from residual stress seems to be the most likely explanation for the lower strength of Al_2O_3/Al_2O_3 joints compared to monolithic Al_2O_3.

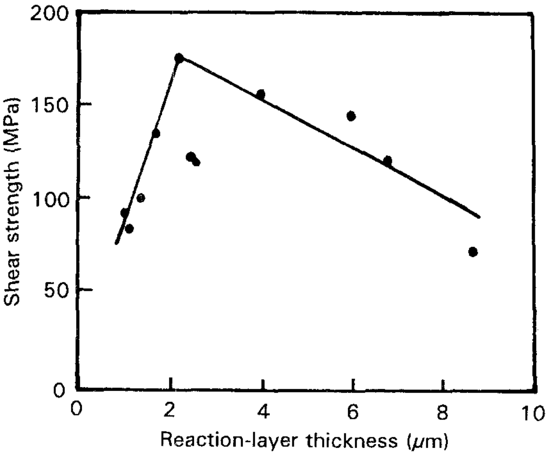

Figure 10 The relationship between the joint strength and the reaction-layer thickness.

In our previous work [11], we also discovered that the effects of the joining temperature and holding time or reaction-layer thickness on the joint strength in ZrO_2/ZrO_2 joints showed the same tendency as that in Al_2O_3/Al_2O_3 joints, and the reaction-layer thickness and properties of the reaction products play an important role in determining the joint strength. The maximum strength value was obtained when the reaction occurred under a temperature slightly higher than the melting point of the filler alloy and with a relatively short reaction time.

Nakao *et al.* [8] have investigated the effect of the reaction layer on bonding strength of Si_3N_4/Mo joints using copper-based active insert metals, and also found the same tendency of the effect of reaction layer on the joint strength, and pointed out that the bonding strength at room temperature could be improved by controlling the reaction-layer thickness to an optimum thickness. An insufficient reaction resulted in a low bonding strength in a thinner reaction layer, but defects in the reaction layer, such as a porous zone and cracks, increased with increasing reaction-layer thickness; they became one of the main causes of the decrease in bonding strength.

Kim *et al.* [6] also reported that in studies of the wetting, reaction and bonding of silicon nitride by Cu–Ti alloys, the shear strength of Si_3N_4 to Si_3N_4 was affected by the morphology and thickness of the reaction layer, rather than the wettability.

Joining pressure affected the interfacial reaction and hence affected the joint strength. When the joining pressure was low, the contact area between the ceramic and the filler metal increased with increasing pressure, and diffusion and reaction occurred easily increasing the joint strength, but when the joining pressure was too high, the joint strength was decreased. This result and the fact that there were areas where the filler metal bled from the joint interface, predicated that the lower strength was due to insufficient interfacial reaction between the ceramic and the filler metal because the titanium content reacting with the ceramic was decreased. Suganuma *et al.* [5] reported that in Si_3N_4 joints, the strength distribution of the samples bonded under 0 MPa pressure was very wide and the Weibull slope was 2.8, while that of the joints bonded at 0.05 MPa pressure was 14.8. In the case of the pressureless bonding experiments, there were many pores and the thickness of the filler metal layer was not uniform. On the other hand, under conditions of 0.05 MPa, intimate contact was produced at the interface and this microstructural difference influenced the strength.

4. Conclusions

The effects of joining conditions, such as joining temperature, pressure and holding time, on the joint strength of alumina-to-alumina ceramic joints were investigated. The joint strength was largely affected by the joining conditions involved; it first increased and then decreased with increasing joining temperature, pressure and holding time and depended mainly on the strength between the interfacial reaction layer and alumina ceramic, and the interfacial reaction-layer thickness, as well as the interface morphology. When, at low temperature and short holding time, the interfacial reaction between ceramic and filler metal was insufficient, both the interfacial bonding strength and the reaction-layer strength were low. Therefore, the joint strength was low. When the joining temperature was higher and the holding time was longer, the reaction-layer strength and interfacial bonding strength between the reaction layer and ceramic decreased, because the amounts of brittle reaction products increased and the true contact area decreased, due to the presence of cracks. Hence, a degradation in joint strength occurred.

A low joining pressure was advantageous to joint strength because the true interface contact area increased, while a higher one was disadvantageous due to loss of filler metal at the interface, resulting in insufficient interfacial reaction.

The maximum joint strength was obtained when the reaction occurred under a suitable temperature, pressure and holding time, and the reaction layer thickness was about 2 μm.

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Received 16 November 1993
and accepted 25 March 1994