Joining of alumina with an alumina–zirconia insert under low mechanical pressure

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Mikinori Hotta [a],[*], Naoki Kondo [a], Hideki Kita [a], Tatsuki Ohji [a], Yasuhisa Izutsu [b], Takashi Arima [b], Yasunori Matsumura [b]

[a] Advanced Manufacturing Research Institute, National Institute of Advanced Industrial Science and Technology (AIST), 2266-98 Shimo-Shidami, Moriyama-ku, Nagoya 463-8560, Japan
[b] Ceramics Division, Mitsui Mining & Smelting Co., Ltd., 3-1 Asamuta-cho, Ohmuta, Fukuoka 836-0817, Japan

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ABSTRACT

Two alumina pipes were joined with an alumina–20 mass% zirconia composite insert for 4 and 8 h at 1650 °C and 1700 °C under a low mechanical pressure of 0.05 MPa. Although plate-like voids were observed along the interfaces in the specimens joined at 1650 °C, such voids were replaced with smaller round pores in specimens bonded at 1700 °C owing to local plastic deformation of the insert at the joint interfaces. In addition, the zirconia grains of the insert were deeply incorporated into alumina grains in the parent alumina at the interfaces at 1700 °C. Because of these microstructural effects, the specimens joined at 1700 °C showed relatively high flexural strengths both at room temperature and at 1200 °C that were close to that of the parent alumina, and most of the specimens were fractured from the parent alumina.

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1. Introduction

Alumina ceramics are most widely used as manufacturing components in various industries because of their relatively low cost and good mechanical/chemical properties. Some alumina components, such as tubes for rotary kilns and stages for liquid crystal supports, are long, typically over 2 m. However, it is difficult to fabricate such long components as single units because huge production facilities are needed and the available forming techniques are limited. An alternative approach is to join several shorter ceramic units to create a long component [1–3]. When employing a joining technique for this purpose, it is essential to join the ceramic units by locally heating the joint region in order to reduce the energy consumption and cost in the production process. Furthermore, ceramic components are most often used in high-temperature and/or corrosive environments, and excellent sealing performance is thus required to prevent gas and liquid leaks. In order to meet these requirements, the joint parts need to have sufficient mechanical, thermal and chemical properties. Furthermore, concerning the joining procedure, the mechanical

pressure externally applied during heat treatment should be as low as possible because joining at high applied pressures is costly.

Many researchers have reported on joining alumina ceramics with various interlayers such as metal, glass, and ceramic [4–16]. Among these methods, solid-state diffusion has an advantage in that the joined alumina has good heat resistance and chemical stability equivalent to those of the parent alumina. However, high mechanical pressure during joining is necessary for enhancing diffusion and achieving strong bonding at the joint interface. Nagano et al. [15] tried to join two similar alumina blocks directly at 1500 °C under an applied pressure of 12.5 MPa without using an insert material. When employing fine-grained alumina (grain size: 0.64 μm), joining was possible, although a number of residual pores were found at the joint interface between the parent alumina blocks. The room-temperature flexural strength of the joined alumina specimen was ∼280 MPa, whereas that of the parent alumina was approximately 400 MPa. However, joining of a coarse-grained alumina (grain size: 5.87 μm) was not successful under this condition. Miyazaki et al. [16] also joined two blocks of alumina (grain size: 4.0 μm) using a pure alumina slurry as an insert material at 1650 °C without substantial external pressure. They found that large voids 10–100 μm wide, as well as many pores approximately 2 μm diameter, were formed in the joint layer, leading to poor sealing performance.

It has been known that alumina–zirconia composites obtained by adding 20 mass% zirconia (3 mol% yttria-doped zirconia,
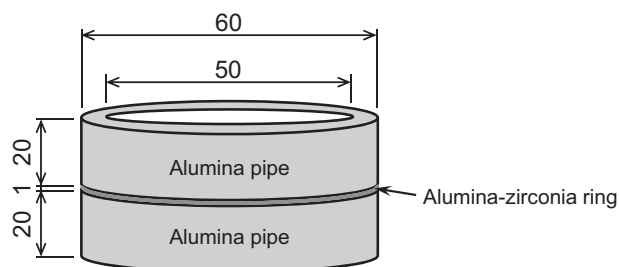
Fig. 1. Schematic illustration of parent alumina and alumina–zirconia insert used for joining.

3Y-ZrO$_2$) have superplastic deformability at high temperatures [17], which effectively makes joining easier by enhancing local deformation and allowing full contact at the joint interfaces [15]. When joining alumina (grain size: 5.87 µm) with an alumina–zirconia composite (grain size: 1.06 µm) at 1500 °C under a mechanical pressure of 12.5 MPa, a joint strength of ∼200 MPa was obtained [15], although such a high mechanical pressure is not suitable for actual industrial applications, as mentioned above. In this study, the joining of coarse-grained alumina pipes using an alumina–zirconia composite (80 mass% alumina and 20 mass% zirconia) as an insert material was attempted under a low mechanical pressure of 0.05 MPa at high temperatures of 1650 °C and 1700 °C.

2. Experimental procedure

Dense, high-purity alumina pipes with a height of 20 mm and outer and inner diameters of 60 and 50 mm, respectively (SSA-S grade, Nikkato Co., Ltd., Osaka, Japan), were used for the parent alumina. The joint surfaces (end faces of the pipes) were ground with a #400 diamond wheel before joining. For the insert material, alumina–zirconia composite rings were fabricated from mixed powders of 80 mass% alumina (Al-160SG-4 grade, Showa Denko K. K., Tokyo, Japan) and 20 mass% zirconia (3 mol% yittria-doped zirconia, TZ-3Y-E grade, Tosoh Corp., Tokyo, Japan). The powder mixing was carried out by ball milling the powders in a plastic pot with alumina balls and ethanol for 48 h. The mixed powder was dried and passed through a sieve. A dispersant (A6114, Toagosei Co., Ltd., Tokyo, Japan) and distilled water were then added to the powder mixture and mixed by ball milling to prepare a slurry for slip casting. The green bodies obtained by slip casting were dried and then sintered at 1600 °C for 2 h in air. Before joining, the joint surfaces of the alumina–zirconia sintered bodies were ground to a height of 1 mm with a #400 diamond wheel. For the joining process, the alumina–zirconia rings with a thickness of 1 mm were placed between the parent alumina pipes. The pipe–ring–pipe structures were then placed under a mechanical pressure of 0.05 MPa and heated at temperatures of 1650 °C and 1700 °C for holding times of 4 and 8 h in air, as shown in Fig. 1. This mechanical pressure was selected by assuming the weight of an alumina pipe 1 m high with outer and inner diameters of 160 and 150 mm, respectively, when it is stood vertically since ceramic pipes of such length are often joined to obtain a longer one [2].

In order to investigate the microstructure of the joint region and measure the joint strength, the joined alumina pipes were cut perpendicular to the joint surface. The surfaces of the obtained specimens were then polished with a 0.5-µm diamond slurry and thermally etched. The etched surfaces were observed by scanning electron microscopy (SEM; JEOL Ltd., Tokyo, Japan) to characterize the microstructure of the joint region. The joint strength of the specimens was measured at room temperature and 1200 °C using a four-point bending method in a universal testing machine (Sintech 10/GL, MTS Systems Corp., USA). The specimens had dimensions
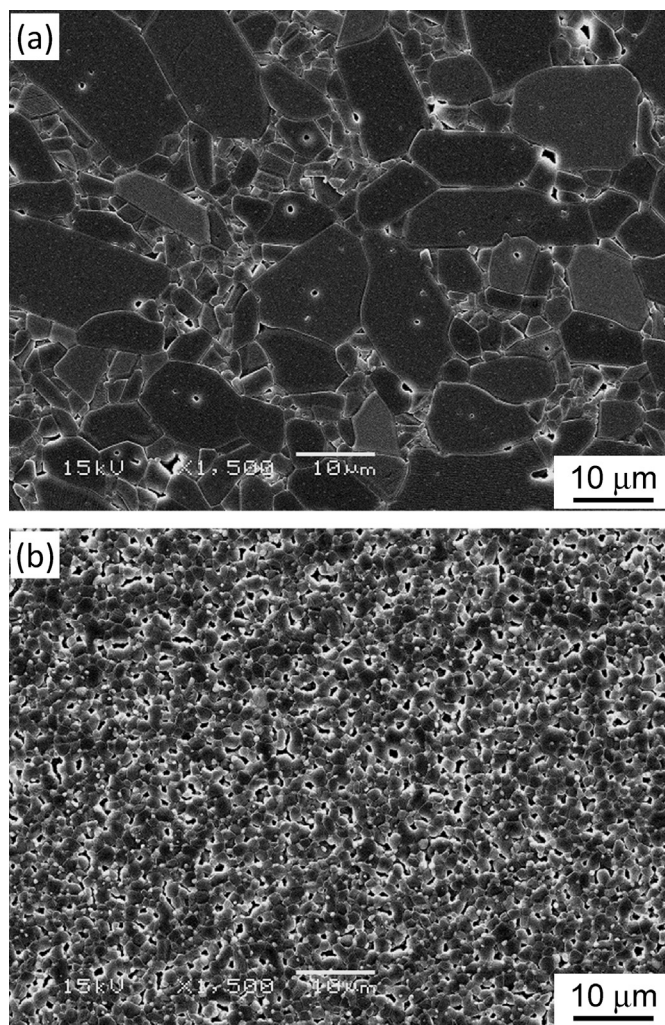

Fig. 2. Scanning electron micrographs of (a) the parent alumina and (b) alumina–zirconia insert before joining.

of 3 mm × 4 mm × 40 mm, and the joint region was placed at the center of the bending bar. The outer and inner spans were 30 and 10 mm, respectively, and the crosshead speed was 0.5 mm/min.

3. Results and discussion

3.1. Microstructure

Fig. 2 shows SEM micrographs of the parent alumina and the alumina–zirconia insert before joining. The alumina grains in the parent alumina were substantially larger than those in the alumina–zirconia insert. The grain sizes of the parent alumina, the insert alumina, and the insert zirconia were 3.0, 1.2, and 0.5 µm, respectively. The relative densities of the parent alumina and alumina–zirconia insert were 99 and 97%, respectively.

Fig. 3a and b shows SEM micrographs of the joint regions of the specimens joined at 1650 °C for 4 and 8 h, respectively, and Fig. 3c and d shows those joined at 1700 °C for 4 and 8 h, respectively. Some plate-like voids whose thickness was typically 1–2 µm were observed along the interfaces for the specimens joined at 1650 °C; such voids were replaced with smaller round pores in specimens bonded at 1700 °C. Cracks due to a thermal expansion mismatch between the parent alumina and the alumina–zirconia insert were not observed in the joint regions. It should also be noted that because of the grain growth, the grain sizes of both the parent
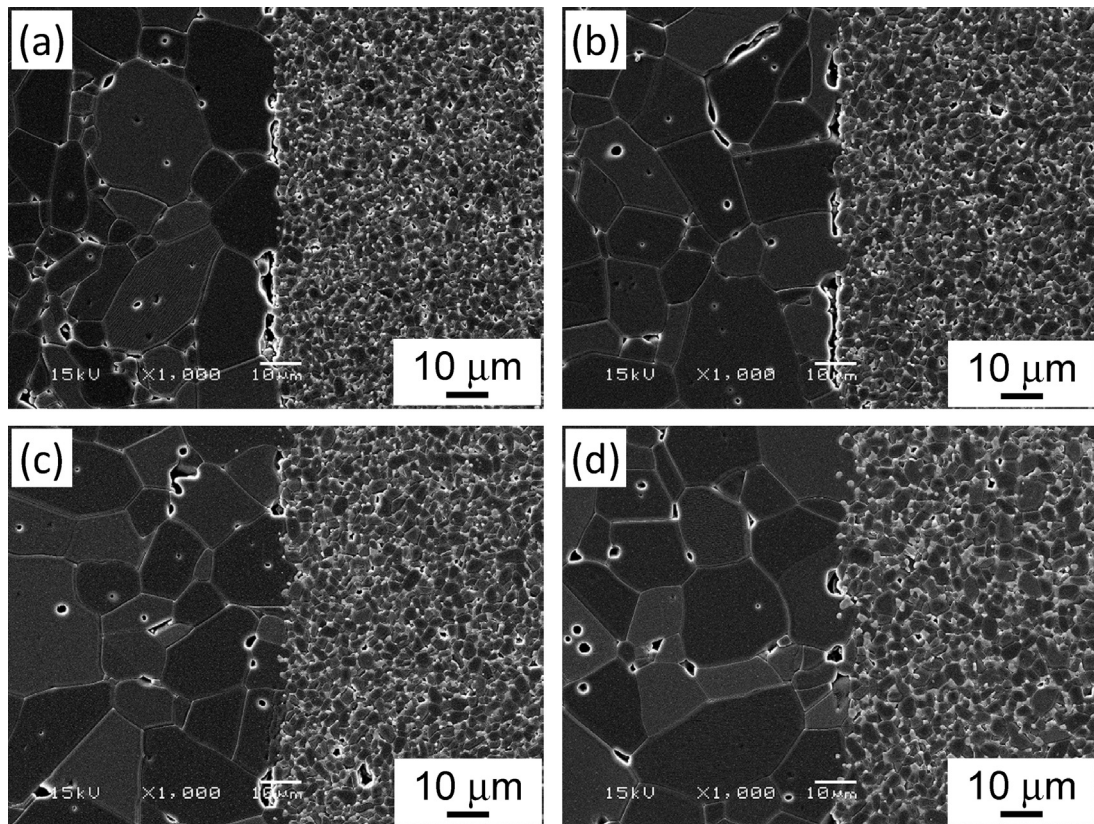
Fig. 3. Scanning electron micrographs of the joint regions of the specimens joined at 1650 °C for (a) 4 h and (b) 8 h and at 1700 °C for (c) 4 h and (d) 8 h with an applied pressure of 0.05 MPa. The left and right sides are the parent alumina and the joint layer, respectively.

alumina and the alumina–zirconia insert increased as the temperature or holding time increased. The respective grain sizes of the parent alumina, the insert alumina, and the insert zirconia were 4.9, 1.4, and 0.7 μm, respectively, at 1650 °C for 4 h, 6.7, 1.8, and 0.8 μm, respectively, at 1650 °C for 8 h, 8.0, 1.5, and 0.8 μm, respectively, at 1700 °C for 4 h, and 8.7, 2.0, and 1.0 μm, respectively, at 1700 °C for 8 h.

Fig. 4a and b shows magnified views of the joint interfaces of the specimens joined at 1650 °C and 1700 °C for 4 h, respectively. The light gray grains are zirconia from the alumina–zirconia joint layer, indicating that zirconia grains were incorporated into alumina grains in the parent alumina at the joint interfaces. This incorporation was enhanced in the specimens joined at 1700 °C. It is assumed that the incorporation was caused by a combination of diffusion of zirconia grains from the joint layer and growth of alumina grains in the parent alumina.

As stated in the introduction, it can be considered that the voids and pores at the joint interfaces were removed by the local plastic deformation of the joint layer (insert material) [15,17,18]. The plastic deformation behavior of the alumina–20 mass% zirconia composite equivalent to the composition in this work has been reported by Wakai et al. [17]. The strain rate of a polycrystalline material is generally expressed by the following equation:

$$\frac{d\varepsilon}{dt} = A\frac{\sigma^n}{D^p}\exp\left(\frac{-Q}{RT}\right)$$

where A is a dimensionless constant, σ the applied stress, D the grain size, n the stress exponent, p the grain-size exponent, Q the activation energy for deformation, and R and T the gas constant and temperature, respectively. Assuming grain-boundary diffusion, p is 3. Q and n were reported to be 750 kJ/mol and 1.9 (the mean), respectively, between 1250 and 1450 °C [17], and it can be assumed

that these deformation parameters did not change substantially in the higher temperature range of 1650–1700 °C because the alumina and zirconia grains in the alumina–zirconia composite did not show significant growth. The strain rate of the alumina–20 mass% zirconia composite (grain size: 0.9 μm) under a stress of 15 MPa at temperatures of 1650 °C and 1700 °C was determined from Wakai's data, which was $\sim 4 \times 10^{-3}$ s^{-1} and $\sim 1.2 \times 10^{-2}$ s^{-1} at 1650 °C and 1700 °C, respectively. When the stress was 0.05 MPa and the average sizes of the alumina and zirconia grains in the composite were 1.05 μm and 1.15 μm at 1650 °C and 1700 °C, respectively, after a holding time of 4 h, the strain rates at those temperatures were $\sim 4 \times 10^{-8}$ s^{-1} and $\sim 1.2 \times 10^{-7}$ s^{-1}, respectively. Hence, the deformation ability of an alumina–zirconia insert whose thickness is 1 mm can be very roughly estimated to be approximately 0.5–1 μm at 1650 °C for 4–8 h and 1.5–3.5 μm at 1700 °C for 4–8 h. It should be noted that owing to the local stress concentration at the joint interfaces, the actual deformability is expected to be higher than that estimated. It is assumed that this difference in deformability between 1650 °C and 1700 °C may have resulted in the 1–2 μm-thick plate-like voids seen at 1650 °C that were scarcely observed at 1700 °C.

3.2. Mechanical strength

Fig. 5 shows the room-temperature flexural strengths of the specimens joined at 1650 °C and 1700 °C for 4 and 8 h under a mechanical pressure of 0.05 MPa, including the numbers of specimens fractured from the parent alumina and the joint interfaces between the parent alumina and the alumina–zirconia joint layer. The strength of the parent alumina before joining and those of specimens heated at 1650 °C and 1700 °C without mechanical pressure are also shown for comparison. The joint strength at 1650 °C
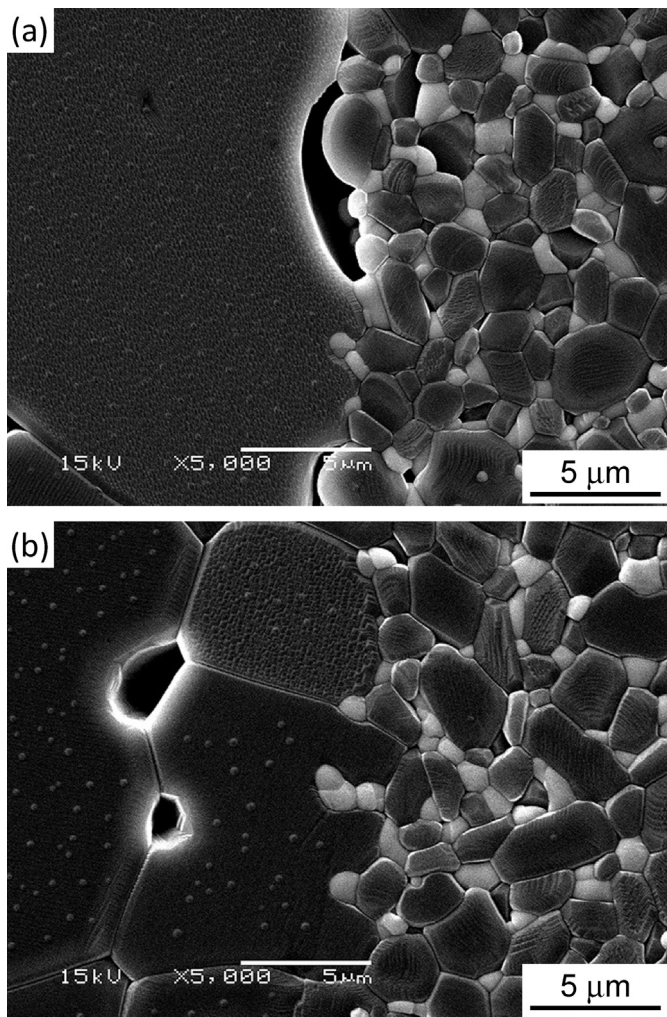
Fig. 4. Scanning electron micrographs of the joint regions of the alumina specimens joined at (a) 1650 °C and (b) 1700 °C for 4 h with an applied pressure of 0.05 MPa. The left and right sides are the parent alumina and the joint layer, respectively.

for 4 h was 166 MPa and that at 1650 °C for 8 h was 185 MPa, and most of the joined specimens were fractured from the joint interfaces. When the joining temperature increased from 1650 °C to 1700 °C for 4 h, the joint strength increased to 224 MPa and that
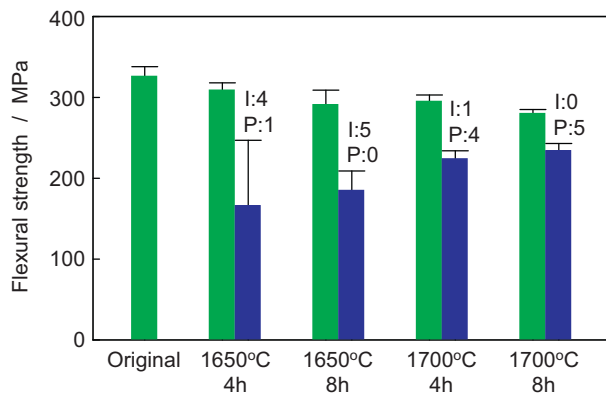

Fig. 5. Room-temperature flexural strengths of the specimens joined at 1650 °C and 1700 °C for 4 and 8 h under a mechanical pressure of 0.05 MPa (blue bar) with the numbers of specimens fractured from the parent (P) and the interface (I). The strengths of the parent alumina before joining and those of specimens heated at 1650 °C and 1700 °C without pressure are also shown for comparison (green bar).
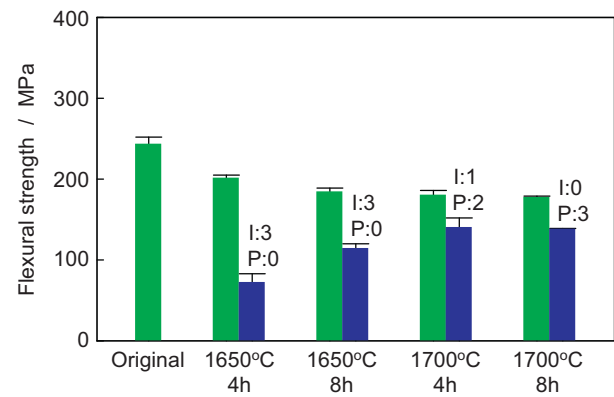

Fig. 6. High-temperature (1200 °C) flexural strengths of the specimens joined at 1650 °C and 1700 °C for 4 and 8 h under a mechanical pressure of 0.05 MPa (blue bar) with the numbers of specimens fractured from the parent (P) and the interface (I). The strengths of the parent alumina before joining and those of specimens heated at 1650 °C and 1700 °C without pressure are also shown for comparison (green bar).

at 1700 °C for 8 h was 234 MPa, and most of the joined specimens were fractured from the parent alumina. The increases in the joint strength are most likely due to the decrease in the number of voids at the interfaces and the incorporation of zirconia grains of the joint layer into alumina grains in the parent alumina, as revealed in Figs. 3 and 4. On the other hand, while the original alumina before joining had an average strength of 326 MPa, the strength after the heat treatment decreased with increasing heating temperature and holding time owing to the growth of alumina grains, as shown in Fig. 3. The strength of the specimens joined at 1700 °C for 8 h was relatively close to that of the parent alumina heated under the same conditions.

Fig. 6 shows the high-temperature (1200 °C) flexural strengths of the specimens joined at 1650 °C and 1700 °C for 4 and 8 h under a mechanical pressure of 0.05 MPa, as well as those of the parent alumina before joining and those of specimens heated at 1650 °C and 1700 °C without pressure. As compared to the room-temperature strengths, all the high-temperature values were somewhat lower owing to softening and sliding of the grain boundaries. The joint strength at 1650 °C for 4 h was 72 MPa and that at 1650 °C for 8 h was 114 MPa, and all of the joined specimens were fractured from the joint interfaces, whereas the joint strength at 1700 °C for 4 h was 140 MPa and that at 1700 °C for 8 h was 138 MPa, and most of the specimens were fractured from the parent alumina, as was the case for the room-temperature strength tests. Similarly, the strength of the heat-treated parent alumina decreased with increasing heating temperature and holding time owing to the growth of alumina grains. The high-temperature strengths of the specimens joined at 1700 °C for 4 h and 8 h were relatively close to those of the parent alumina heated under the same conditions.

Here we compare the current results with those of similar previous studies on the joining of alumina ceramics. Nagano et al. [15] succeeded in joining two blocks of fine-grained alumina without an insert material at 1500 °C under an applied pressure of 12.5 MPa, whereas joining was not successful for the case of coarse-grained alumina whose grain size was similar to that in this study. Miyazaki et al. [16] joined coarse-grained alumina ceramics using a pure alumina slurry as an insert material at 1650 °C without substantial external pressure, resulting in a joint strength of 283 ± 50 MPa at room temperature, which was a little higher than the strength obtained in this study (234 ± 9 MPa). However, they also reported that several large voids 10–100 μm wide were observed in the joint layer, leading to very wide scattering of strength values and poor sealing performance. Esposito et al. [11] joined alumina blocks with an insert of CaO–Al$_2$O$_3$–SiO$_2$ glass at a low temperature of 1250 °C

without applied pressure, and they obtained a joint strength of 230 MPa, which is similar to that found in this work. However, owing to softening of the glassy phase in the joint layer, the high-temperature mechanical properties would degrade substantially as compared to the alumina joint fabricated in this study.

4. Conclusions

High-purity alumina pipes were joined with a dense alumina–20 mass% zirconia composite insert sintered for 4 and 8 h at 1650 °C and 1700 °C under a low mechanical pressure of 0.05 MPa. This mechanical pressure was selected by assuming the weight of an alumina pipe 1 m high with outer and inner diameters of 160 mm and 150 mm, respectively, when it is stood vertically. Some plate-like voids were observed along the interfaces in the specimens joined at 1650 °C, whereas such voids were replaced with smaller round pores in specimens bonded at 1700 °C because of local plastic deformation of the insert at the joint interfaces. In addition, the zirconia grains of the insert were deeply incorporated into alumina grains in the parent alumina at the interfaces at 1700 °C. Because of these microstructural effects, the specimens joined at 1700 °C showed relatively high flexural strengths both at room temperature and at 1200 °C. The flexural strength values were close to those of the parent alumina, and most of the specimens were fractured from the parent alumina. This study indicates that high-strength alumina joints could be obtained without high mechanical pressure by forming a strong bond at the joint interfaces with the use of an alumina–zirconia insert.

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