Microwave joining of alumina with alumina/zirconia insert under low pressure and high temperature

Naoki KONDO,1 Mikinori HOTTA, Hideki HYUGA, Kiyoshi HIRAO and Hideki KITA

National Institute of Advanced Industrial Science and Technology (AIST), 2266–98 Shimo-shidami, Moriyama-ku, Nagoya 463–8560, Japan

Joining of alumina with an alumina/zirconia insert by microwave heating was carried out at a low mechanical pressure of 0.17 MPa and high temperatures of 1650–1750°C. Joining occurred by diffusion bonding at 1650 and 1700°C, which are lower than the eutectic temperature of alumina/zirconia, and by liquid-phase assisted bonding at 1750°C, which is higher than the eutectic temperature of alumina/zirconia. Alumina joining under a low mechanical pressure and high temperature occurred successfully. Alumina joined at 1700 and 1750°C showed strengths of 310 and 212 MPa, respectively. Finally, joining by microwave local heating was demonstrated.

Key-words : Alumina, Joining, Microwave, Temperature, Pressure, Strength

1. Introduction

Alumina (Al2O3) is an important engineering ceramic and it is widely used in industry as balls, saggars, tubes, plates, beams, and so on. Industry is now using large alumina components such as long tubes for rotary kilns, and long stages and beams for liquid crystal support. The tubes, stages, and beams can now be as long as 3 m.3) Much longer components are required for industrial applications, but making single components is difficult because of limitations on production techniques and facilities.

Direct joining of alumina is preferred because the components often require high temperature strength, stiffness, or corrosion resistance. If the use of inserts for joining is unavoidable, they must be composed of alumina or an alumina-based composite. Many joining techniques have been studied in an attempt to satisfy this requirement.

Diffusion bonding is a major method for directly joining an alumina part (purity >99.5%) with another part. The joining mechanism for diffusion bonding has been studied widely.2) Direct joining requires high temperatures and mechanical pressure. Alumina joined at 1500°C and 12.5 MPa showed strength of ~280 MPa.5) A composite made by adding 20 wt% zirconia (3 mol% yttria-doped zirconia, 3Y-ZrO2) effectively makes joining easier by enhancing local deformation and allowing contact between parts at the interface. A composite joined at 1500°C and 12.5 MPa showed high bending strength of ~500 MPa.5)

Liquid phase formation is another way to join alumina, and zirconia is a candidate for this process. The alumina–zirconia system forms a eutectic liquid phase above 1710°C.4) The formed liquid phase is expected to enhance local deformation and diffusion for joining.

From the viewpoint of industrial facilities, many industrial furnaces operate at elevated temperatures of up to 1800°C. However, few furnaces, except for hot-press furnaces, contain devices for applying high mechanical pressure. Therefore, high temperatures of up to 1800°C are acceptable, whereas low mechanical pressure comparable to that actualized by applying weight on the component is preferred for making long components by joining.

For joining, heating is required only near the joint. Therefore, local heating around the joint is preferred to conserve energy. Microwave heating has the potential to locally heat the area around the joint.8) Such heating requires 1/10th the electric power of conventional resistance heating required for the joining of silicon nitride.9) Thus, microwave heating was used in this work.

Considering the above circumstances, joining of alumina under low pressure and high temperature was attempted in this paper. To actualize the joining, an alumina/zirconia composite was chosen as an insert since it seemed favorable to enhance diffusion bonding or liquid phase enhanced bonding. Microwave heating was used to examine an ability of local heating. The micro-structures and mechanical properties of joined alumina were investigated after the joining procedures, and the possibility of the joining under low pressure and high temperature was discussed.

2. Experimental

The bulk alumina bodies used in this study were fabricated from high-purity (>99.5%) alumina powder (Al2O3, AL160SG4 by Showa Denko K.K.); dense alumina-sintered bodies of size 30 × 10 × 20 mm were prepared by sintering the powder at 1600°C for 2 h. The surfaces (30 × 10 mm) used for joining were machined with a #200 whetstone.

An alumina/zirconia composite plate insert was fabricated from the powders of alumina (TM-DAR by Taiimei Chemicals Co. Ltd.) and zirconia (3Y-ZrO2, TZ-3Y-E by Tosoh Corp.). A dense plate of size 30 × 10 × 1 mm was prepared by sintering the alumina–20 wt% zirconia mixed powder at 1600°C for 2 h. The surfaces (30 × 10 mm) used for joining were machined with a #200 whetstone.

Alumina/zirconia composite plates were used as microwave susceptors, and alumina fiberboard was used as the insulator. Alumina bodies and alumina fiberboard are poor microwave absorbers at low temperatures. On the other hand, the zirconia...
granules in the alumina/zirconia susceptor are good microwave absorbers and can be heated by microwave radiation.

Schematic diagram of the arrangement of alumina bodies, alumina/zirconia insert, susceptor, insulator, and so on, are shown in Fig. 1. The alumina/zirconia insert was placed between two alumina bodies to make a joint. The joint plane was horizontally aligned. Four side (vertical) planes of the bodies were surrounded by the susceptor plates. This configuration was placed inside the alumina fiberboard, which in turn was placed in a microwave furnace. A low uniaxial mechanical pressure of 0.17 MPa was applied to the joint plane by placing an alumina rod, steel rod, and steel weight on the alumina bodies.

A microwave furnace with four magnetron sources [frequency: 2.45 GHz, maximum output: 1.5 kW × 4 (6.0 kW in total)] was used for heating. The temperature inside the susceptor plates (without alumina bodies) was measured using thermocouples up to 1500°C, and that outside the plates was measured using a pyrometer. The temperature higher than 1500°C was measured only by pyrometer. The temperature inside the susceptor plates was extrapolated from the difference between the temperatures measured by the thermocouples and the pyrometer.

The configuration of alumina bodies was heated up to 1650, 1700, and 1750°C for 1 h by controlling the microwave output to avoid thermal shock fractures. It was then soaked at these temperatures for 30 min. Hereafter, the specimens of the bodies jointed at 1650, 1700, and 1750°C will be referred to as “L”, “M”, and “H”, respectively.

Specimens for microstructure observations and strength measurements were cut from the joined alumina body. Microstructure observations were performed by scanning electron microscopy (SEM) and energy dispersive X-ray spectrometry (EDX). Specimens of size 3 × 4 × 40 mm were prepared for strength measurements. These specimens had a joint at the center of a bend bar. Their four-point bending strength was measured in accordance with the JIS R1601 standard using outer and inner spans of 30 and 10 mm, respectively, and at a displacement rate of 0.5 mm min⁻¹.

3. Results and discussion

3.1 Microstructure

Figure 2 shows SEM micrographs of the joined specimen around the joint. The zirconia grains appear bright in the micrograph. The top and bottom sides were alumina/zirconia joint and alumina bulk, respectively. The L specimen contained some connected pores at the interface between the alumina bulk and alumina/zirconia joint. At the temperature and pressure for the L specimen, deformation and diffusion were insufficient for the elimination of pores. On the other hand, in the M specimen, the connected pores were not observed, and only a few isolated pores were found. Grains in the alumina bulk were slightly coarsened compared to those in the L specimen by heating. The grain sizes of alumina bulk were the same regardless of the distance from the interface. No zirconia was found in the alumina bulk. The observed microstructures were similar to those previously reported, therefore, joining in the L and M specimens occurred under a diffusion bonding condition.

As the elimination of pores at the interface was mainly occurred by the local deformation around the interface, effect of temperature on local deformation of alumina/zirconia insert is discussed here. Deformation behavior of alumina-20 wt% zirconia composite was reported. Strain rate of the composite was proportional to exp(Q/RT). Here Q is an apparent activation energy for deformation, and it was reported to be 750 kJ/mol between 1250 and 1450°C. R and T are gas constant and temperature, respectively. Assuming Q was similar up to 1700°C since alumina and zirconia grains were still fine, strain rate became three times faster when the temperature increased from 1650 to 1700°C. Therefore, the higher temperature effectively decreased the number of pores at the interface by enhancing local deformation.

The microstructure of the H specimen was substantially different from that of the L and M specimens. The H specimen contained few pores at the interface. Grains in the alumina/zirconia joint as well as in the alumina bulk near the joint were substantially coarsened. Zirconia was found within the coarsened alumina grains in the joint as well as at alumina grain junctions in the alumina bulk. Figure 3 shows the microstructures of the H specimen, which shows alumina grains around and away from the joint. The grains around the joint and those located

---

Fig. 1. Schematic diagram of the arrangement of alumina bodies, alumina/zirconia insert, susceptor, insulator, and so on.

Fig. 2. SEM micrograph of the region around the joint interface. L, M and H were joined at 1650, 1700, and 1750°C, respectively. The top and bottom sides are the alumina/zirconia joint and alumina bulk, respectively. The joint interfaces between the joint and bulk are horizontal in the middle of the micrographs.
1.25 mm from the joint interface were larger than, but those located 2.5 mm from the joint were the same to, those farther in the bulk. The diffusion of zirconia into alumina bulk, which initially occurred within the alumina/zirconia insert, was examined by SEM. Figure 4 shows a typical zirconia particle existing at the junction of alumina grains. This micrograph shows a position ~1.25 mm from the joint interface. The amount of zirconia reduced as the position goes farther from the joint. A very small number of zirconia particles was found even ~5 mm from the joint interface.

A liquid phase was formed during joining because the joining temperature of the H specimen was higher than the eutectic temperature. The liquid phase contributed to joining by enhancing the growth of alumina grains in and around the joint and the diffusion of zirconia into alumina bulk. Therefore, joining in the H specimen occurred under a liquid-phase assisted bonding condition. A typical lamellar eutectic microstructure was not observed. This is probably due to the lower amount of zirconia, i.e., 20 wt%, which was about half the eutectic composition, 42.6 wt%.

3.2 Strength

The strengths of the joined specimens at room temperature (R.T.) and elevated temperatures are shown in Fig. 5. Typical fractured specimens at R.T. are shown in Fig. 6. The strength of an original specimen (before joining) was measured for reference, and it was found to be 447 MPa.

The L specimen had strength of 186 MPa at R.T. All the specimens were fractured from the joined interface between the alumina/zirconia joint and alumina bulk. Fracture occurred from the pores because connected pores existed at the interface in this specimen. Strength measurement at elevated temperatures was not performed because of the existence of connected pores.

The M specimen showed the highest strength among the three specimens, with strength of 310 MPa at R.T. All specimens were fractured from the alumina bulk; therefore, a few isolated pores did not act as fracture origin. The strength of this specimen was remarkably lower than that of the original specimen (447 MPa). Coarsened grains are known to be disadvantageous for achieving high alumina strength.11) The high joining temperature effectively eliminated pores at the joint interface under low mechanical pressure; however, it caused grain growth of the alumina bulk and led to strength degradation. The strength of the M specimen was maintained up to 800°C. It slightly decreased above 800°C, and was 259 MPa at 1200°C. Most of the specimens were fractured from the alumina bulk at elevated temperatures; ~1/4 of the specimens were fractured at the joint interface. These specimens fractured at the interface had strengths comparable to those fractured from the alumina bulk; therefore, the joined interface was sufficiently strong even at elevated temperatures.

The H specimen had strengths of 212 and 169 MPa at R.T. and 1200°C, respectively. These strengths are significantly lower than those of the M specimen. All the specimens were fractured from
bulk alumina at R.T. and at elevated temperatures. Thus, the joining itself occurred well. The strength of the alumina bulk was largely degraded because the alumina bulk grains were remarkably coarsened by heating at high temperatures with eutectic liquid phase formation. This is the reason for fracturing from the alumina bulk.

4. Joining by local microwave heating

Joining of alumina by microwave local heating was attempted with reference to a previous study on the joining of silicon nitride by microwave local heating. Two high-purity alumina tubes, 25 (outer) and 20 (inner) mm in diameter and 200 mm in length, were used. An alumina/zirconia insert ring, 1 mm in thickness, was placed between the two tubes. An alumina/zirconia composite susceptor cylinder, 40 mm in height, was placed around the joint. Thus, the section near the joint was locally heated by microwave radiation. Joining was carried out under a mechanical pressure of 0.22 MPa at 1700°C for 30 min. Joining of alumina tubes by microwave local heating occurred successfully, as shown in Fig. 7.

5. Conclusions

The alumina bulk and alumina/zirconia insert combination was joined at a low mechanical pressure of 0.17 MPa and high temperatures of 1650–1750°C. Microwave heating was used for joining.

Joining was conducted by diffusion bonding at 1650 and 1700°C, which are lower than the eutectic temperature of alumina/zirconia. The specimen joined at 1650°C contained only a few isolated pores. The specimen joined at 1700°C had strength of 310 MPa at room temperature. Fracture occurred from the alumina bulk and not from the joined interface. Thus, joining occurred successful at 1700°C.

Joining also occurred by liquid-phase assisted bonding at 1750°C, which is higher than the eutectic temperature of alumina/zirconia. Few pores were found at the joint interface. As a result of the liquid phase formation, alumina grains in the alumina/zirconia joint and in the alumina bulk near the joint were substantially coarsened. The specimen had strength of 212 MPa at room temperature and fracturing occurred from the bulk alumina. The degraded strength in comparison to that joined at 1700°C was the result of the coarsening of grains, although the joining itself occurred well and a joint interface with few pores was formed.

Finally, joining by microwave local heating was successfully demonstrated.

Acknowledgement This work was supported by METI and NEDO, Japan, as part of the project, “Innovative Development of Ceramics Production Technology for Energy Saving”.

References

4) Phase Equilibria Diagrams, Fig. 4377, American Ceramic Soc.