Joining of silicon nitride by microwave local heating

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Microwave local heating was used to join silicon nitride. Glass was placed at the joint between two silicon nitride pipes and a silicon carbide susceptor was placed around the joint. The susceptor was locally heated by absorbing the microwave radiation. On heating to 1500°C, the glass melted and filled the space between the pipes, thereby joining the two pipes. Average strength of the specimen cut out from the joined pipe was 446 MPa. This demonstrates the potential of microwave local heating for joining silicon nitride.

Key-words : Microwave heating, Local heating, Susceptor, Silicon nitride, Glass, Strength

1. Introduction

Silicon nitride ceramics are widely used for industrial components in various manufacturing industries, such as stalk tubes, ladles, and heating element protection tubes in the aluminum casting industry and transfer rolls in the steel industry. Such components are frequently large and sometimes exceed 1 m in length. The industries now require large-sized, light-weighted and/or complex-shaped components. As the silicon nitride components are generally fabricated as single units, the following three problems are encountered. 1) Large production facilities are required to manufacture large components. 2) Green machining (i.e., machining of compacted powder prior to sintering) to form thin walls for lightweight or complex-shaped components is difficult. 3) Handling and sintering are difficult due to the own weight of the components. Consequently, conventional fabrication procedures are reaching their physical limits.

One technique that has the potential to overcome such problems is joining. By fabricating small ceramic parts and joining them, large-sized, light-weighted and/or complex-shaped components are expected to be realized. Thus, a lot of researches on joining, not only for silicon nitride but also for other ceramics, have been performed from the past to the present.1)–4) It is preferable to join silicon nitride using simple equipment and reducing energy consumption during processing. Microwave heating appears to have the potential to satisfy these requirements. However, only a few reports on joining of ceramics by microwave heating have been published.1)–3)

Microwave heating is a unique heating technique in that it heats materials (including ceramics) directly and volumetrically; thus, it can realize rapid, uniform heating. This is in contrast to conventional heating techniques in which the surface of a body is initially heated by radiation and heat is subsequently conducted to the center of the body. Additionally, microwave heating requires only a simple chamber consisting of stainless-steel plates, a magnetron, and an insulator. This simple equipment is also the advantage of microwave heating.

Microwave absorption for heating is depending on material specifics. This means that local heating can be achieved by using an appropriate combination of materials. For joining, heating is only required near the joint. Thus, microwave local heating appears to be a promising technique for joining.

This study demonstrates the ability of microwave local heating to join silicon nitride pipes. A combination of silicon nitride (poor microwave absorber at low temperatures), alumina fiber-board insulator (poor microwave absorber), and silicon carbide susceptor (microwave absorber) was used. The temperature distribution was measured to confirm that local heating was achieved. Silicon nitride pipes were then heated to 1500°C to melt glass (which was used as an insert) and to join the silicon nitride pipes. Microstructural observations and strength measurements were carried out to investigate the properties of the joined pipes.

2. Experimental procedure

Figure 1 shows a schematic drawing of the arrangement used for microwave local heating. Silicon nitride pipes with outer and inner diameters of 28 and 16 mm, respectively, were used for joining. This silicon nitride is a commercially available thermocouple protection tube. Super Sialon (Hitachi Metals, Ltd.), for aluminum casting. Two pipes of 90 and 60 mm in length were used as the upper and lower parts. Glass was used as an insert; it was prepared by mixing raw powders, $\alpha$-Si$_3$N$_4$ (SN-E10, Ube Industries Ltd.), Y$_2$O$_3$ (Shin-Etsu Chemical Co. Ltd.), Al$_2$O$_3$ (AL16O5G4, Showa Denko K.K.), and SiO$_2$ (Kojundo Chemical Lab. Co., Ltd.). The composition of the mixed powder was 30.1 mass % Si$_3$N$_4$–43.4 mass % Y$_2$O$_3$–1.8 mass % Al$_2$O$_3$–

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Fig. 1. Schematic diagram of the arrangement of silicon nitride pipes, insulator, and susceptor.
14.7 mass % SiO₂, following a previous study. The glass powder was placed between the upper and lower pipes to form a joint.

Alumina fiberboard with a hole of 30 mm in diameter was used as the insulator. The silicon nitride pipes were placed inside the hole. Silicon carbide (SiC) granules were used as the susceptor. The heating zone is expected to be very short using the susceptor. The granules were placed around the joint with a thickness of 5 mm and a length of 40 mm (i.e., 20 mm above and below the joint). Silicon nitride and alumina insulator are poor microwave absorbers, making them difficult to heat by microwave irradiation. In contrast, SiC readily absorbs microwaves, causing it to heat up. This difference in microwave absorptions is expected to generate a temperature distribution, resulting in local heating.

The temperature was measured using a pyrometer by observing the SiC granules through a hole in the insulator. In addition, K-type thermocouples were inserted in the silicon nitride pipes to ensure that local heating conditions existed up to 1150°C.

A microwave furnace with a magnetron (frequency: 2.45 GHz; maximum output power: 6 kW) was used for heating. Heating was performed in N₂ gas flow.

Specimens for microstructural observations and strength measurements were cut from the joined silicon nitride pipes. Microstructure observations were performed by optical microscopy (OM) and scanning electron microscopy (SEM). Seven specimens (3 × 4 × 40 mm) were prepared for strength measurements; these specimens have a joint at the center of a bend bar and the tensile direction was perpendicular to the joint. Fourpoint bending strength was measured according to DIN 1601 using outer and inner spans of 30 and 10 mm, and a displacement rate of 0.5 mm/min.

3. Results and discussion

3.1 Microwave local heating

Temperature measurements were first performed along the length direction of the silicon nitride pipe to determine the temperature distribution. Temperatures at the center of the pipe (i.e., at the joint) and at 3 and 6 cm above the center were measured. Figure 2 shows the results. The temperature was measured up to 1150°C, since the thermocouple melted at higher temperatures. When the temperature at the center reached to 1100°C, the temperature at 6 cm above the center was about 250°C lower than that at the center. This demonstrates that local heating around the SiC susceptor was successfully achieved. Assuming that the temperature difference between the center and at 6 cm above the center increases continuously, it will reach 350°C when the temperature at the center is 1500°C.

Figure 3 shows the heating profile for joining. To reduce thermal shock within the materials due to rapid heating, the microwave power was gradually increased so that the joint temperature increased to 1500°C in 40 min. After heating for 10 min at 1500°C, the microwave power was turned off and the joined pipe was cooled. Despite the almost constant microwave radiation power, the temperature increased steeply above 1300°C during heating. It has been reported that microwave absorption of liquid-phase sintered silicon nitrides increases sharply when the temperature exceeds the temperature at which the grain-boundary glassy phase starts to soften. Therefore, it is considered that, in addition to the heat generated by the SiC susceptor, the heat generated by microwave irradiation of silicon nitride itself plays an important role above 1300°C.

Figure 4 shows images of joined silicon nitride pipes. It reveals that the two pipes were successfully joined by glass. A cross-sectional image of the joint is also shown. Softened glass filled the gap between the pipes well. This demonstrates that microwave local heating is a promising technique for joining silicon nitride.

3.2 Microstructure and strength of joined silicon nitride

Figure 5 shows a SEM micrograph of the area around the
joint. The glass joint was about 170 μm thick (see Fig. 5(a)). No separation between the silicon nitride and the glass joint is visible. A crack perpendicular to the joint was also observed. The edge of the crack extended into the silicon nitride (see Fig. 5(b)). Similar cracks due to the thermal expansion mismatch between silicon nitride and glass have been reported. Figure 5(d) shows the microstructure of the glass joint. No elongated silicon nitride grains were observed. Many pores with diameters of ~0.5 μm were observed. These pores were due to α-silicon nitride grains, which were removed by etching; the α-silicon nitride grains did not dissolve into the glass due to the relatively low joining temperature (1500°C) and the short joining time (10 min).

The strength of specimens that had been cut from the joined pipes was 446 ± 35 MPa (average strength and standard deviation). Fracture was occurred at an interface between the silicon nitride and the glass joint. In contrast, the strength of specimens cut from the original pipe was 780 MPa. Therefore, the strength of the joined specimen was about 60% that of the initial specimen.

Silicon nitride joined by glass with the same composition exhibited a strength of ~80 MPa (joined without pressure) and ~550 MPa (joined with an applied mechanical pressure). Most silicon nitrides joined by glass with no or low pressure (<0.01 MPa) exhibited strengths in the range 400–600 MPa, whereas silicon nitride joined by glass under optimized conditions had a strength of over 900 MPa. Therefore, the strength obtained in this study (446 MPa) is comparable to the moderate results previously reported. Therefore, microwave local heating can achieve a joint strength comparable to those obtained by conventional resistance furnace heating.

4. Summary

Microwave local heating was used to join two silicon nitride pipes.

1) Local heating was achieved by placing silicon carbide granules around the joint as the susceptor. When the temperature at the center of the silicon nitride pipe is 1100°C, the temperature at 6 cm above the center is about 250°C lower than that at the center.
2) Heating was performed up to 1500°C and silicon nitride pipes were successfully joined by glass. Softened glass filled the gap between the pipes.
3) The average strength of specimens cut from the joined pipe was 446 MPa. This is comparable to the moderate strengths obtained by conventional resistance furnace heating in previous studies.

These results demonstrate that microwave local heating is a promising technique for joining silicon nitride.

Microwave local heating is a promising technique for joining silicon nitride parts to fabricate large and/or complex-shaped ceramic components. Further optimization of the joining conditions and investigation of joined specimens are currently in progress.

Acknowledgement This research was supported by METI and NEDO, Japan, as part of the Project for the Development of Innovative Ceramics Manufacturing Technologies for Energy Saving.

References


