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Sintering of alumina by 2.45 GHz microwave heating

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Abstract

Sintering behavior of alumina by 2.45 GHz microwave heating was investigated on the base of an isothermal barrier structure, which consisted of an inner blanket (composed of mullite and α-alumina) and an outer one (fibrous alumina-silica board). The temperature difference within the sample was predicted from the densities of samples with different sizes, i.e. different volume to surface ratios. The temperature difference was largest at about 1500°C. The thermal stresses caused by the temperature difference did not produce microcracks in the sample. Very little temperature difference in the sample would exist at 1600°C. The homogeneous microstructures were observed by SEM in the sample sintered at 1600°C. Bending strength was measured for test pieces, which were cut from the center of sample (size before sintenng: 60 × 60 × 30 mm) sintered at 1600°C.

Keywords: Al_2O_3; Grain size; Insulators; Mechanical properties; Microwave processing; Sintering

1. Introduction

Microwave heating has been applied to various ceramics, such as Al_2O_3[1,2], ZrO_2,[3] Si_3N_4[4] and PZT[5] for rapid heating and improvement of microstructures of ceramics. However, many studies have shown local heating, which is the strong temperature dependence on dielectric loss of most ceramics for 2.45 GHz microwave. Local heating generally leads to thermal stresses and cracks in the ceramics. Therefore, the size of sintering ceramics by 2.45 GHz microwave had been limited to several centimeters. We developed an idea "isothermal barrier" to inhibit heat transfer through the surface of the sample in our previous studies[6,7] of 2.45 GHz microwave sintering of porcelain. The structure of the isothermal barrier consists of a thin inner most layer with the identical microwave characteristics to the sample and thick outer layers with low heat conductivity and low microwave absorption. The isothermal barrier was effective in creating the uniform temperature distribution in porcelain products. If the large sizes of advanced ceramics with homogeneous microstructure are obtainable by 2.45 GHz microwave sintering based on the isothermal barrier, this new process will contribute to the economical fabrication for the industries.

In this present study, sintering of alumina was studied by using 2.45 GHz microwave heating with the isothermal barrier at various temperatures. The temperature difference within the sample was predicted from the densities of samples with different sizes, i.e. different volume to surface ratios. We discussed the effect of microwave heating on the sintering behavior of alumina in the final stage of sintering.

2. Experimental procedures

2.1. Sample preparation

As a starting material, high purity Al_2O_3 powder (AL-160-SG4, Showa Denko Co. Japan) with an average particle size of 0.6 μm was used. The alumina powder was wet-ball milled with 0.6 mass% dispersant (D-305, Chukyo Yushi Co. Japan), 2 mass% binder (WA-310, Mitsui Chemical Co. Japan) and suitable amount of water for 28 h to eliminate agglomerate, and then the slip was de-aired. Green bodies were formed by slip casting into a plate with the size of 200 × 200 × 30 mm and were calcined at 1050 °C for 1 h by an electric furnace. Test pieces with various dimensions were cut from the calcined bodies.

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2.2. Microwave sintering

The samples were sintered by using 2.45 GHz microwave kiln, which had multimode cavity (0.4m^3) equipped with two rotating fans and four magnetron microwave generators. Three generators could be controlled the output power from 0 to 1.5 kW continuously, the other was fixed at 2.5 kW. The samples were put in the isothermal barrier, which was made of the double layer structure with inner blankets and outer blankets, as shown in Fig. 1. The plate with 5 mm in thickness (bulk density 2.28 g cm^{-3}), composed of mullite and α-alumina, was used for inner blanket material. Fibrous alumina-silica board of 40 mm in thickness was used for the outer blanket as insulator. The inside dimensions of the inner blanket were 180 × 180 × 95 mm. Microwave heating was performed up to maximum temperature with a maximum power (7 kW). The temperature inside the surface of the inner blanket was measured by the infrared pyrometer. Typical microwave heating profile is shown in Fig. 2.

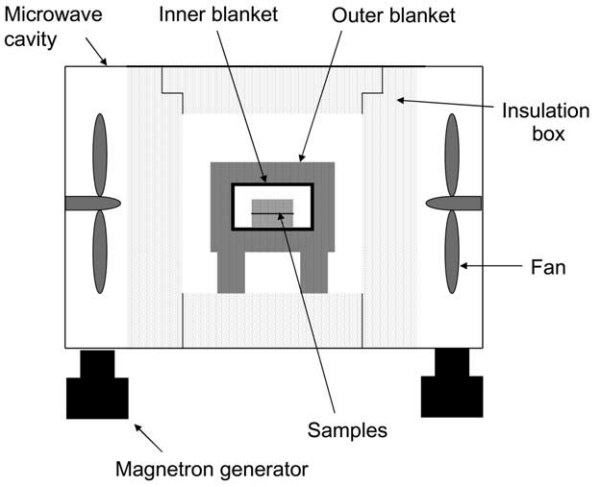

Fig. 1. Microwave kiln with isothermal barrier structure.

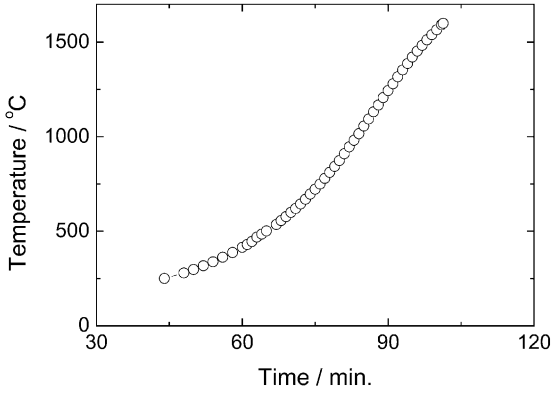

Fig. 2. Typical microwave heating profile.

2.3. Measurement

Densities of sintered samples were measured by the Archimedean method. Bending strength was determined by the three-point bending test in accordance with JIS R-1601. Microstructures were observed by the scanning electron microscope (SEM)(S-2400 Hitachi, Japan) on the polished and thermal-etched cross sections of sintered samples cut by a diamond wheel.

3. Results and discussion

3.1. Microwave heating behaviors

During heating, the interior of the sample is heated directly by microwave. As a result of heat valance between the microwave power absorbed within the sample and the rate of energy loss from the sample surface, the temperature gradient generally exists in the sample. In case of the microwave heating with the isothermal barrier, the temperature gradient in the sample during sintering should not theoretically exist. However, it is very difficult to achieve this condition exactly. The temperature gradient can be accepted if it will not reduce the properties of the sintered sample. The heating behaviors were investigated in the sample of different volume to surface ratios. Because microwave energy absorption is proportional to the volume of the sample and heat loss is proportional to the surface area of the sample, the temperature gradient in the sample should be dependent on the volume to surface ratio.

The densification of the samples with the different volume to surface (V/S) ratios against the sintering temperature is shown in Fig. 3. The sizes of the used samples were 30 × 30 × 2 mm, 30 × 30 × 5 mm, 30 × 30 × 10 mm and 30 × 30 × 30 mm, and then the V/S ratios were 0.05, 0.19, 0.3 and 0.5, respectively. The relative densities of the samples (calcined alumina bodies) before sintering were 62%. The densities of the samples decreased with the increase of V/S ratio at $<$1400°C. The densities became approximately equal for the sample with less than 0.3 V/S ratios at $>$ 1400°C. The sample with the highest V/S (0.5) ratio showed densities about 3% higher than others in the range of 1450–1550°C. The density became approximately equal at $>$ 1600°C. The densification behaviors of the samples varied depending on the V/S ratios.

The densification at 1400°C in the samples with the different V/S ratios against the holding time is shown in Fig. 4. The densities of all these samples abruptly increased in a short time of 15 min, especially, the increase in density of the sample with the 0.5 V/S ratio was remarkable. These densities slowly increased with the increase of the holding time, and similar densification curves were shown for the samples with the less

than 0.3 V/S ratios. The sample with the 0.5 V/S ratio maintained higher densities than the other ones through the holding time up to 120 min. The density difference between the sample with the 0.5 V/S ratio and 0.05, 0.19 and 0.3 V/S ratio slightly decreased, despite extending the holding time up to 120 min. In the case of conventional heating, the center of sample is heated by conduction of heat from the sample surface. Therefore, in spite of the V/S ratios, the densities in the samples will become equal by holding the maximum temperature long.

3.2. Estimation of temperature gradient in alumina sample

The samples with the V/S ratio under 0.3 (10 mm in thick) showed almost the same densification curves above 80% of the theoretical density as shown in Figs. 3 and 4. These results indicated the homogenous temperature distribution in the sample. This means that the temperature distribution in the sample with the ratio under 0.3 (10 mm in thick) was homogeneous between the surface and the center. The density of the surface was assumed to be the same as the sample with 0.05 V/S ratios. The density of the interior was calculated by Eq. (1).

$$\rho_{0.5} \times V_{0.5} = \rho_{0.05} \times V_s + \rho_c \times V_r \quad (1)$$

where, $\rho_{0.5}$: measured density of the sample with 0.5 V/S ratio; $V_{0.5}$: whole volume of the sample with 0.5 V/S ratio; $\rho_{0.05}$: measured density of the sample with 0.05 V/S ratio; V_s: volume between the surface and 5 mm in deep of the sample with 0.5 V/S ratio; ρ_c: calculated density of the residual volume of the sample with 0.5 V/S ratio.

V_r: $V_{0.5} - Vs$

The calculated densities in the center and those of the surface (V/S:0.5) were shown in Fig. 5. The density difference in the sample reached a maximum at 1500°C on the surface. When the surface temperature is 1500°C, the temperature at the interior can be estimated to be about 1600°C from the low densification curves in the Fig. 5. The temperature difference in the sample would become larger as the increase of the V/S ratios. The microwave sintering experiments were performed with a large sample of 60 × 60 × 30 mm in size (V/S ratio: 0.75). The densities at the center and the ends were measured with the test piece of 5 mm in thick cut from the sample. The density changes of the sample sintered at 1400, 1500 and 1600°C were shown in Fig. 6. The density at the center became largest with the sample sintered at 1500°C, and this result agreed with the curves shown in Fig. 5. No visible cracks were observed in the sample. The grain growth differences were not observed between the center and the surface of the sample sintered at 1600°C with the 0.5 V/S ratio, shown in Fig. 7. Large bending strength 568 MPa was obtained

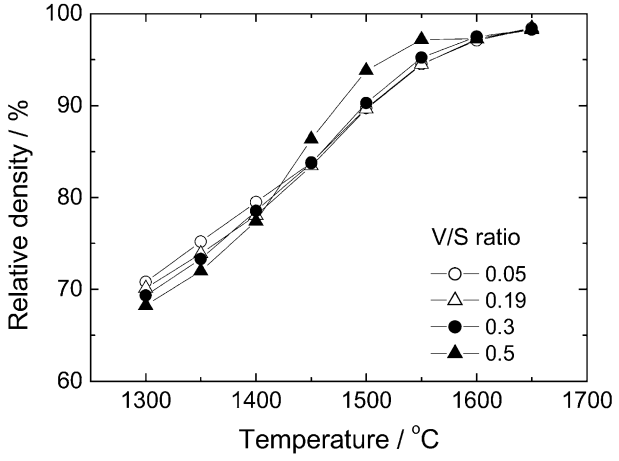

Fig. 3. Densification of the samples with the different volume to surface ratios against the sintering temperature.

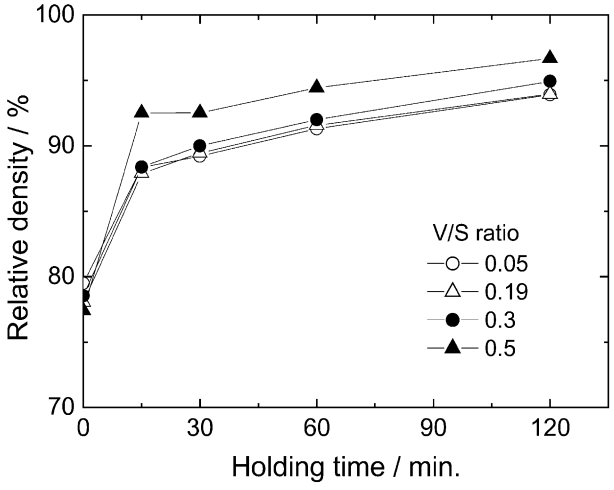

Fig. 4. Densification at 1400 °C in the samples with the different volume to surface ratios against the holding time.

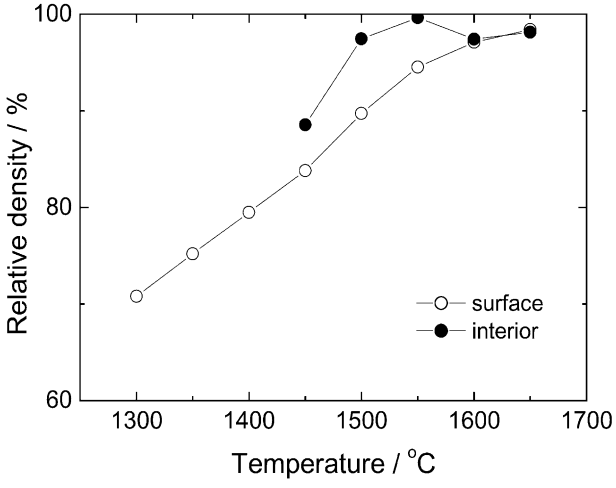

Fig. 5. Calculated densities in the center and those of the surface (V/S: 0.5) against the heating temperature.

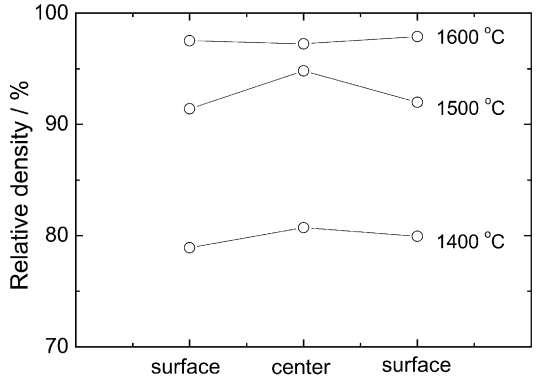

Fig. 6. Density variations in the sample sintered at 1400, 1500 and 1600°C.

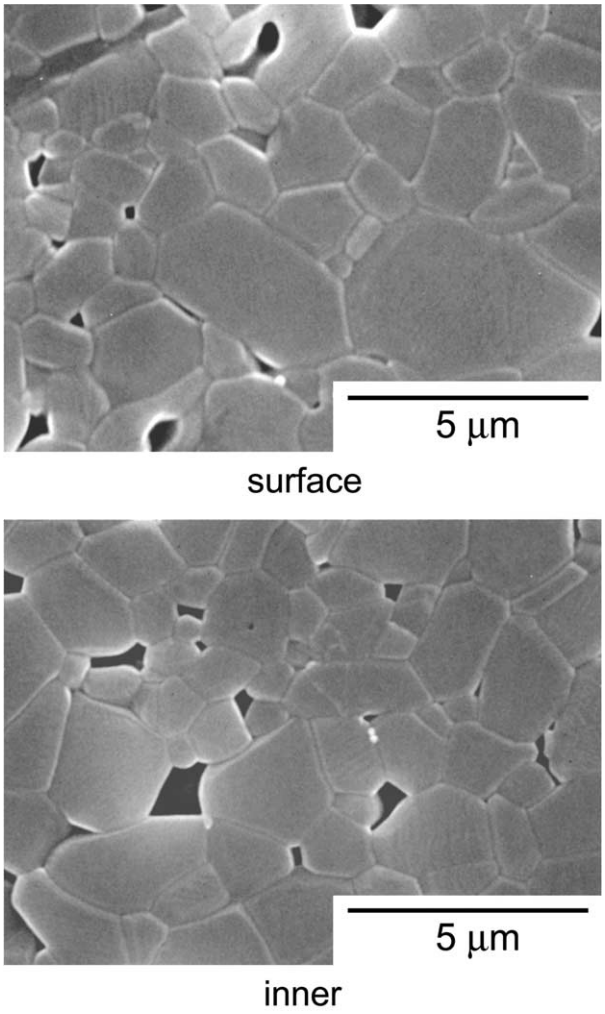

Fig. 7. SEM photographs of the surface and the center of the sample sintered at 1600°C.

with the test piece cut from the center of the sample with 0.75 ratio. These results indicated that the local microwave heating did not arise in the center of the sample. It is well known that the alumina sample absorbs more microwave as the temperature is higher. The temperature difference in the sample will be larger as the sintering temperature is higher. As a result, the sample may be rupture by the local microwave heating. Microwave absorption characteristics vary depending on the microstructures, which change on heating sample prepared from alumina powder. We considered the sample absorbs more microwave on the rapid densification. In the final stage of sintering, the temperature distribution in the sample became homogenous because the surface absorbs much microwave than the interior. This study enabled the microwave sintering of alumina sample with the ratio up to 0.75 by using the inner blanket made of mullite and α-alumina.

For the sintering sample with a large ratio of V/S, the inner blanket material, where it is possible to raise the temperature at high speed, could be used. In the temperatures above 1600°C, the sample is sintered at a small heating rate for homogeneous temperature distribution by heat conduction.

4. Conclusion

A sintering experiment of alumina sample by 2.45 GHz microwave heating based on the isothermal barrier was performed. Density measurement after sintering in samples with different volume to surface ratios showed that the densification of the sample was influenced by the volume to surface ratio. The temperature difference in the sample was estimated from the difference between the surface (measured) and the interior (calculated). Though large temperature differences existed at temperatures near 1500°C, the temperature differences became smaller as the temperature reached 1600°C. This result was also supported by SEM observation, in which the same grain size was obtained with the surface and the interior of the sample. Alumina sample with sizes of 60 × 60 × 30 mm was sintered to more than 98% of theoretical density by rapid heating up to 1600°C. The large bending strength 568 MPa was obtained. Thermal runaway was avoided by using the isothermal barrier structure.

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