High Frequency Microwave Sintering of Alumina Ceramics

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Abstract: - Processing alumina by using a very high microwave frequency, 300 GHz to produce a high density compact was performed. A high purity alpha alumina powder was used as starting material and a high power 300 GHz gyrotron material processing system was used for microwave sintering. A starting compact was prepared by slip casting and followed by cold isostatic pressing (CIP) to increase sample density before sintering. Examination on sintered samples revealed that an alumina compact with density up to 99.1 % of theoretical density (TD) and grain size less than 1 micrometer can be produced through 300 GHz sintering. Compared to conventional results, the densification of 300 GHz at same sintering temperature is much higher and the grains are also finer. The higher in densification observed in 300 GHz sintered alumina could be attributed to an increase in mass transport rate during sintering. This could be associated to the increase in either driving force or apparent activation energy of diffusion. The finer grains of 300 GHz sintered samples can be attributed to difference in time consumption for sintering between the sintering methods. The enhanced densification rate with low grain growth in 300 GHz sintering found in this study suggests that a high density with fine-grained alumina ceramic can be produced by using this microwave frequency.

Key-Words: - Alumina, high frequency microwave sintering, densification, grain size, apparent activation energy

1 Introduction

Sintering ceramics by using microwave energy not only opens the possibilities for more homogeneous processing but also possible to processing the ceramic with low cost and get better properties. Alumina is one of the most sintered ceramics by microwaves [1]. The previous reports in microwave sintering of alumina ceramic suggested that the microwave effect on alumina densification increases with an increase of microwave frequency [2-4]. It suggested that it is possible to produce a higher density alumina compact by using a higher microwave frequency. That is useful for alumina application. However, despite the potential implication of higher frequency microwave processing for the alumina ceramic application, there have been little published work which using microwave frequency higher than 83 GHz [4]. Microwave, 2.45 GHz has been the most commonly used microwave frequency because of its availability and feasibility. For ceramic processing, 2.45 GHz systems with a few kilowatts of power are also widely available. However, several problems such as inhomogeneous and thermal run-away in the heating of materials with hot spots come inherent in 2.45 GHz microwave heating. By operating at a higher frequency, the wavelength becomes shorter and inhomogenity became less difficult problem [5]. However, the high frequency microwave material processing systems were not widely available. Recently, an advanced progress in the development of electromagnetic radiation sources, such as gyrotrons, has opened the possibility for developing a material processing materials with higher frequency electromagnetic waves, such as, millimeter and sub-millimeter waves [5-8]. Moreover, some other advantages such as higher heating efficiency even for poor dielectric loss material can be achieved.

Recently, processing ceramics by using a very high microwave frequency, in sub-millimeter wave range, 300 GHz by using a Sub-millimeter wave Material Processing System (SMMW-MPS) in FIR Center University of Fukui have been reported. Various kinds of ceramics such as zirconia and silica as well as alumina have been succesfully sintered [9-12]. Some enhancement because of microwave effect were revealed. For alumina ceramic, the enhancement of densification compared to the conventionally sintered samples was found and reported by authors [12-14]. However, the maximum density acheived after sintered by 300 GHz was only 96 % of TD. Moreover, the mechanism of the 300 GHz waves enhanced alumina sintering is not fully understandable yet.

In this work, alumina green samples were prepared by cold isostatic pressing method before processing by using 300 GHz. Because microwave sintering is sensitive to sample before sintering, there are may a higher density compact can be acheived. Moreover, for more understading the mechanism, the examination of grain growth of sintered samples were also performed. Results of conventional sintering were also described for comparison.

2 Material and Method

Sample Preparation. A high purity alumina powder AES-11C (Sumitomo Chemical Co. Ltd., 99.8 %, 0.4 μm average particle sizes, and 7-8 m^2/g specific surface area) was prepared as the starting material. The powder was dispersed in de-ionized water with 0.3wt% dispersant, an ammonium polycarboxylate acid to form a slurry. The dispersant was added to the slurry to prevent the agglomeration of the suspended alumina particles. The slurry was ball milled in an alumina jar for 22 hours to remove hard agglomerates prior to compaction. A cylindrical sample with a diameter of 20 mm, and a height of 5 mm was formed by slip casting. This sample size was expected to provide enough penetration depth of 300 GHz waves. After formed, the samples were pressed by CIP method at 150 MPa and followed by pre-sintered at temperature 600 ^{o}C for 2 hours. The samples after pre-sintered were referred as green samples.

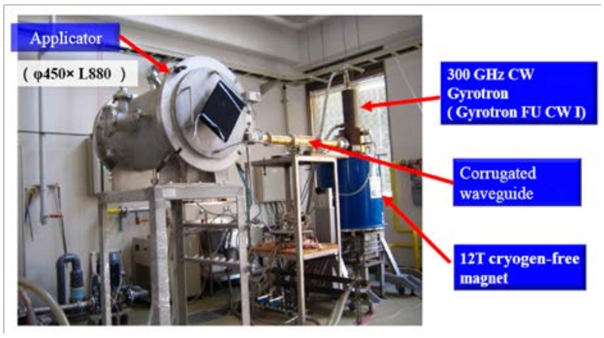

Fig. 1 A sub-millimeter wave material processing system (SMMW-MPS)

Sintering. After pre-sintering, alumina compacts were then sintered by using the SMMW-MPS in FIR Center University of Fukui. The whole sintering system is shown in Fig. 1. A 300 GHz gyrotron installed in liquid helium free 12 T super conducting magnet was applied as radiation source. The system has been applied for material processing system with an output power of 1.75 kW [8]. The microwave generated by the gyrotron is transmitted to the applicator through a low-loss corrugated circular waveguide. The material processing chamber (applicator) has a cylindrical shape with a diameter of 450 mm, and a height of 880 mm (see Fig. 1). It is very large in size compared to the wave length of 300 GHz radiation. The temperature was measured by using Pt-R-type thermocouple which can measure a maximum temperature of over 1700 ^{o}C. In this work, the temperature was measured carefully, and has been tested in several experiments [8,9,12]. In order to achieve the homogenous heating, the sample was placed inside a hollow cylindrical alumina fiber-board (DENKA-ALCEN) as the thermal insulation. Because of the use of thermal insulation, we can assume that no significant temperature difference between the surface and internal sample occurred in this study. After holding the samples at the desired temperature, cooling was carried out naturally after the gyrotron radiation was turned off, with the samples left inside the applicator. The conventional sintering was performed by using an electric furnace (KBF-314N1, KOYO Thermo system). The sintering was performed at high ramp rates (45 ^{o}C/min) for 300 GHz and 2 ^{o}C/min for conventional method to reach

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the targeted temperatures as quickly as possible and to maintain the isothermal conditions. The sintering temperatures were from a temperature of 700 ^{o}C to 1700 ^{o}C in air with holding time of 20 minutes.

Density Measurement. Density of the samples was measured based on the Archimedes`s method where de-ionized water used as an immersion medium. The procedure used in this study was concomitant with the standard test method described in detail by the American Society for Testing and Material Specification, ASTM C373-88 [15]. The density (ρ_b) was calculated as follows:

$$\rho_b = \frac{W_d}{W_{sat} - W_{sus}} \rho_w \qquad (1)$$

where w_d is the weight of the dried sample, w_{sat} is the weight of the sample when with the open pores filled with water (saturated), w_{sus} is the weight of the saturated samples when it is suspended in the water, ρ_w is the density of the water.

Particle Size Calculation. Fracture surfaces were prepared by fracturing the sintered alumina samples. They were then coated with evaporated platinum (Pt) by using an ion sputtering HITACHI E-1030 before SEM photos were taken. The SEM photos were taken every 500 μm from the surface to the center of the samples horizontally by using a scanning electron microscope (SEM) HITACHI S-2600 HS at a magnification of 10.000 times. Gray scale SEM photos were processed by various images processing programs before the grain size calculation. The average grain size was determined from the grain size distribution histogram based on the Gaussian fitting function.

3 Results and Discussion

Green Samples. Because microwave sintering strongly depends on green sample properties, density and microstructure of samples before sintering should be well characterized. Examination of average density of samples before CIPed (un-CIPed samples) is 55.5 % of TD. By using CIP`s pressure, the density averagely increased 2.8 % to 58.3 %. Microstructure characterization was performed from SEM photographs of the fracture surfaces. Fig. 2 shows the SEM photographs of the fracture surfaces of green samples before and after CIP. The photographs show that there are differences in particle arrangement and pore distribution inside the bodies. The CIPed sample has more grain contacts. There also show that the CIPed sample has fewer and less pores than the un-CIPed one. The quantitative analysis of microstructure was performed for calculating the grain sizes.

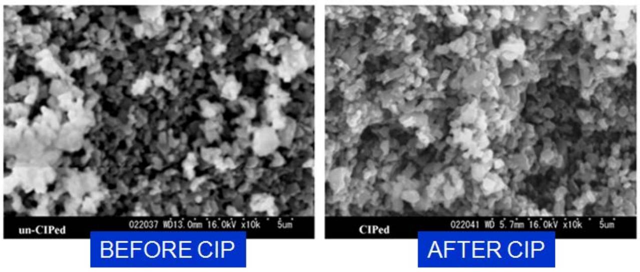

Fig. 2 SEM photograph of the fracture surface of un-CIPed and CIPed green samples

Effect of High Frequency Microwaves on Densification. Fig. 3 shows densification of sintered alumina with increase in sintering temperature for CIPed alumina. The graph shows that alumina ompacts with density up to 99.1 % of TD can be produced through 300 GHz sintering.

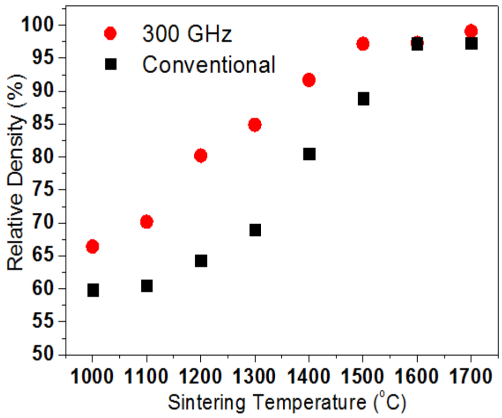

Fig. 3 Density-temperature profile of pressed alumina sintered by different sintering method.

Compared to the conventional sintering, samples sintered in 300 GHz showed a more rapid densification. The greatest shift in densification is 16 % at sintering temperature 1300 °C. It is much higher than that of un-CIPed sample case which only 9 % reported previously [10]. That means, the microwave effect of 300 GHz was greater on CIPed alumina. The higher in densification observed in 300 GHz sintered alumina could be attributed to an increase in mass transport rate during sintering. This could be associated to the increase in either driving force or apparent activation energy of diffusion. An experiment to estimate driving force of sintering is not easy to perform because the force is dependent on not only the density and grain size but also the electric field in the materials. The preliminary estimation of apparent activation energy revealed that the apparent activation energy of 300 GHz sintering is much lower than that in conventional sintering for un-CIPed samples. The details of experiment results of apparent activation energy estimation will be published elsewhere in separate paper. The lower activation energy values suggested that diffusion was faster during sintering which led to the higher densification. The difference in grain sizes between 300 GHz heating with that the conventional even faster diffusion during 300 GHz sintering may be due to the difference in sintering time. Time consumption in conventional heating was much longer than that in 300 GHz. To achieve a sintering temperature of 1500 °C, for example, 300 GHz processing needed only 33.3 minutes, while the conventional method needed 750 minutes, almost 23 times as long as that in SMMW as shown in Table 2. As a result of fast heating in 300 GHz, the grain coarsening did not have time to form even mass transport in 300 GHz sintering was faster than that in conventional heating. A similar phenomenon was reported by Coble for producing transparent alumina by using MgO in which an enhanced densification rate relative to the rate of normal grain growth was observed [19-20].

4 Conclusion

Processing alumina ceramic by using a very high microwave frequency, 300 GHz, to produce a high density and fine grain alumina compact was succesfully performed. A cold isostatic pressing (CIP) applied to increase sample density before sintering was effective for increasing the final density of alumina after sintered by 300 GHz. An alumina compact with density up to 99.1 % of theoretical density (TD) with grain size less than 1 micrometer can be produced. Compared to conventional results, the densification of 300 GHz at same sintering temperature is much higher and the grains are also finer. The results found in this study suggest that a high density with fine-grained alumina ceramic can be produced by using this microwave frequency.

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