

Title	Application of Microwave Thermal Plasma to Ceramics Sintering(Physics, Process, Instruments & Measurements)
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Citation	Transactions of JWRI. 1993, 22(1), p. 31-35
Version Type	VoR
URL	https://doi.org/10.18910/6183
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Application of Microwave Thermal Plasma to Ceramics Sintering[†]

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Abstract

Sintering of pure and MgO-doped Al₂O₃ ceramics is studied using a high-power 915MHz microwave plasma in N₂ gas flow at an atmospheric pressure. For prevention of cracking, green-body samples with initial density of 55% of theoretical were heat-treated in fiber-ceramics liner loaded in the plasma flow before sintering process. Efficient and rapid densification has been obtained within 10 minutes in an N₂ plasma at an input microwave power of 6kW up to final densities higher than 95% of theoretical. Characterization of sintered specimens is discussed in terms of grain growth, hardness, XRD and EPMA analysis.

KEY WORDS: (Ceramic Sintering) (Microwave Thermal Plasma) (Al₂O₃) (MgO-doped Al₂O₃)

1. Introduction

Recently plasma sintering¹⁻²⁾ and microwave or millimeter-wave sintering³⁻⁴⁾ have attracted great attentions as new candidates for the fabrication of fine ceramics, where the following advantages are potentially expected over conventional sintering technique in absence of the intense radiation field and a nonplasma environment:

- (i) An extremely rapid rate of heating and densification of ceramic samples can be achieved.
- (ii) The sintered materials have finer grain size than those obtained with conventional method since the sintering time can be considerably shortened.
- (iii) The sintered fine ceramic materials may attain improved mechanical properties.

We are now studying these new types of sintering processes extensively. In the millimeter-wave sintering experiment using a pulsed high power 60GHz wave, we have succeeded⁵⁾ in the rapid sintering of pure Al₂O₃ from green density with 54% theoretical density (TD) to higher than 99% TD without suffering appreciable grain growth. While on the plasma sintering we are interested in applying an atmospheric-pressure microwave plasma in which a pumping system is not necessary. A high-power microwave at 915MHz was applied to obtain a large volume plasma at atmospheric pressure. In this study results of the plasma sintering experiments with this system on Al₂O₃-based ceramics are described.

2. Experimental Procedure

Figure 1 shows the schematic diagram of the microwave plasma sintering apparatus. A high-power CW microwave at a frequency of 915MHz is transmitted from the left of the figure through a rectangular waveguide (WRJ-1) with TE₁₀ mode. The microwave power can be varied between 0.5 to 20kW. Plasma production is performed in a rectangular waveguide of 180 x 60 x 340 mm. A quartz pipe of 40mm in diameter is inserted into holes on the center of the upper and lower walls of the waveguide at a position of 1/4 wavelength of the 915MHz wave from the end of the waveguide. Ignition of the plasma is given with a vertically movable tungsten stick in an atmospheric pressure rotating flow of N₂ gas around the inner surface of the pipe. The rotating gas flow is effective in cooling the inner wall of the quartz pipe as well as in the stabilization of the plasma flow. Lower part of the quartz pipe is expanded to a diameter of 110mm and connected to a water-cooled stainless steel pipe. Specimens to be sintered are set on the sample holder (made of boron nitride) in this region.

Ceramic powders sintered in this study are high-purity (> 99.995%) α-Al₂O₃ (AKP-50, Sumitomo) and 0.25-wt% MgO-doped Al₂O₃. Figure 2 shows the measured distribution of grain size of AKP-50. As a result the average grain size was approximately 0.2μm. The chemical composition of the AKP-50 is summarized in Table 1. The high-purity and MgO-doped powders were pressed in a uniaxial die at 378MPa for 1.5 min to form green

[†] Received on Aug. 5, 1993

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Transactions of JWRI is published by Welding Research Institute, Osaka University, Ibaraki, Osaka 567, Japan

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compacts (10mm in diameter and 3mm in thickness). Relative densities of these green bodies were 55% TD for pure Al_2O_3 and 54% TD for MgO-doped Al_2O_3 .

As for sintering procedure we have applied two steps to prevent cracking of the samples by the high temperature plasma loading. On the first step we covered the green body with commercially available Al_2O_3 fibers (Al_2O_3 95% + SiO_2 5%) and put them into a short Al_2O_3 tube (purity higher than 99%) of 12mm inner-diameter, after which it was set on the top of the sample holder. The sample was loaded by the plasma at a position of about 10cm below the center of the plasma column. It was heated efficiently without cracking and this procedure corresponds to the presintering of the sample to a desired density by several minutes loading. The second step corresponds to the main sintering, where the presintered sample is heated directly by the plasma flow on the top of the sample holder. The sample density was measured from the shrinkage ratio and the initial density.

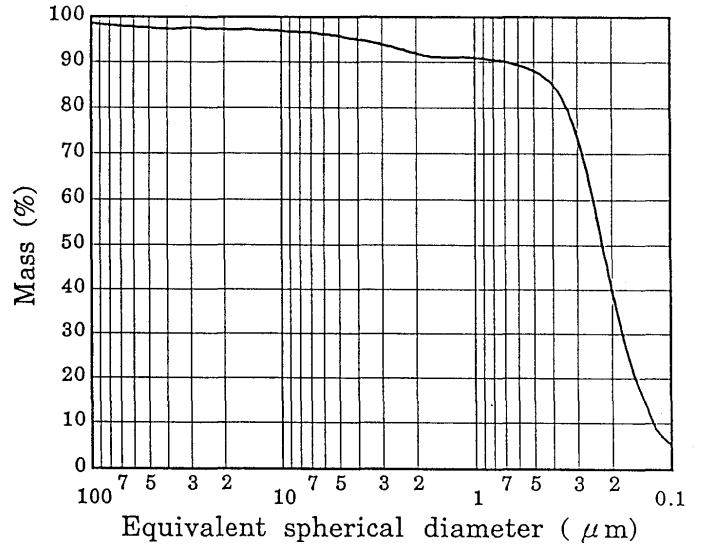


Fig. 2. Grain-size distribution of pure Al_2O_3 (AKP-50, Sumitomo).

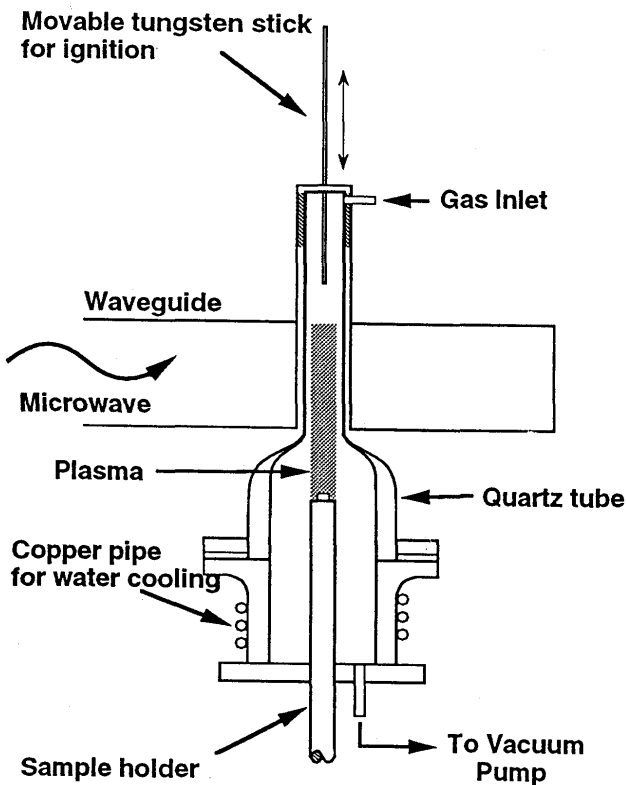


Fig. 1. Schematic of the experimental apparatus.

Table 1. Chemical composition of pure Al_2O_3 (AKP-50, Sumitomo).

Al_2O_3	>99.995 %
Si	<25 ppm
Na	<10 ppm
Mg	<10 ppm
Fe	<20 ppm
Cu	<10 ppm
Average grain size	
0.2 μm	

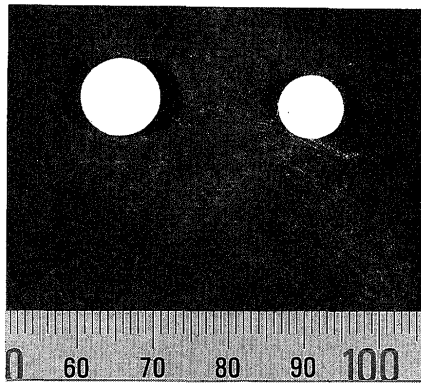


Fig. 3. Pure Al_2O_3 samples; green body (left) and sintered workpiece (right).

3. Results and Discussion

Microwave plasmas in N_2 gas were typically operated at microwave powers ranging from 3kW to 8kW with a gas flow rate of 40l/s at an atmospheric pressure. Below the 3-kW microwave power plasmas were not sustained steadily, while they were not stable when the gas flow rate was lower than 40 l/s. Typical plasma temperature and density in N_2 gas have been determined from spectroscopic measurements and found⁶⁾ to be about 6×10^3 K and $1 \times 10^{14} \text{ cm}^{-3}$, respectively. The plasma dimension increased with the input power and was about 10mm in diameter and 30cm in length at a power of 5kW.

Figure 3 shows an example photograph of the pure Al_2O_3 sample after the main sintering, together with the green sample for comparison. The shrinkage ratio of the sintered sample to the green body (55% TD) confirms that the two-step sintering procedure is successful in obtaining the sintered workpieces approaching 100% TD without appreciable crack formation.

Figure 4 shows the variation of relative density and temperature of sintered Al_2O_3 specimens as a function of sintering time for the case of N_2 plasma produced with a microwave power of 6kW. The samples were presintered to a relative density of 84% TD. We can clearly find that the densification is manifested promptly by the direct loading of the plasma and the density reaches to a value of over 90% TD after 60s from the start of the main sintering. Difference of the final density between pure and MgO-doped Al_2O_3 indicates that the latter has a higher value with faster sintering rate. MgO-doped samples have a density of nearly 100% TD at a time of 300s, while the pure sample attains only a value of about 98% TD even after 1200s. The sample temperature increases rapidly with time and reaches to a value of about 1600°C after about 60s. This result corresponds well with the observation of rapid sintering of the specimen within 60s

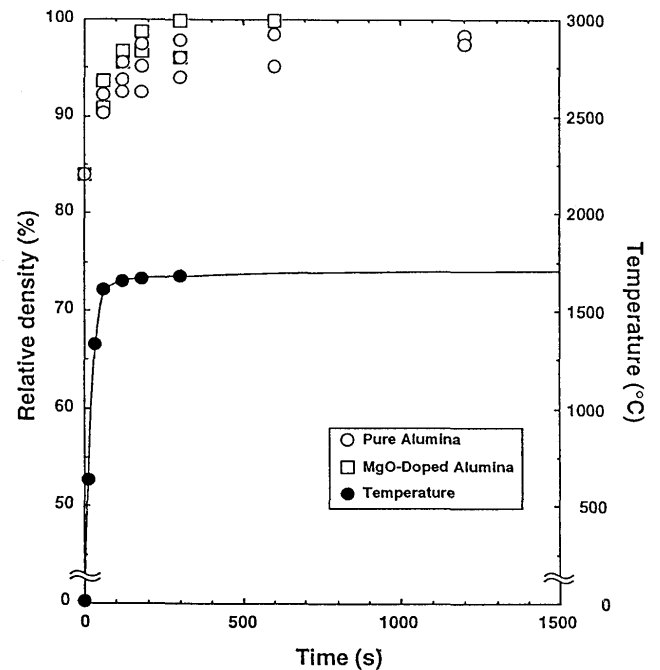


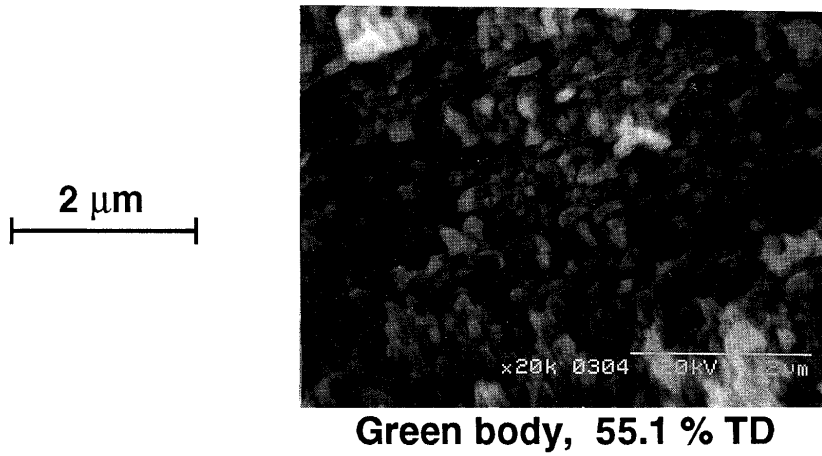
Fig.4. Variations of relative density and temperature of pure and MgO-doped samples as a function of sintering time.

and indicates a close correlation between the sintering temperature and the densification process.

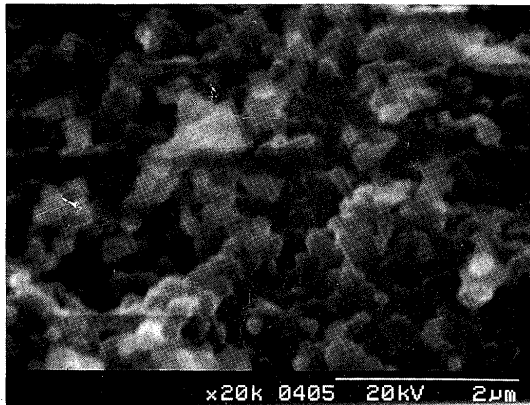
Figure 5 shows the temporal variation of the microstructures in the inner part and on the outer surface of the sintered pure Al_2O_3 samples heated with the N_2 plasma at a microwave power of 6kW. On the top of the figure the green body of 55.1% TD is shown, in the middle the fracture surfaces at the inner part of the samples sintered for 60s (90.2% TD) and 1200s (98.4% TD) are given, and the bottom figures show their outer surface structure exposed to the plasma flow. The grain size of the green body is found to be about 0.1-0.3 μm corresponding well to the grain size measurement shown in Fig. 2. It is interesting that a remarkable difference is observed in the grain growth between the surface and the inner part of the sample. In the inner part the grain size is as small as 0.6 μm , even at 98.4% densification. While on the surface the considerable grain growth is observed to be $>2\mu\text{m}$ at the same density. This is because the plasma heat is supplied to the sample from the surface through heat conduction even when the heating rate is very high.

For the characterization of mechanical property of the sintered samples obtained with the N_2 plasma at a microwave power of 6kW, Vickers hardness was measured at a load of 1.96N and the result is shown in Fig. 6 as a function of the sintering time. The data measured on the outer surface and on the polished surface of the inner

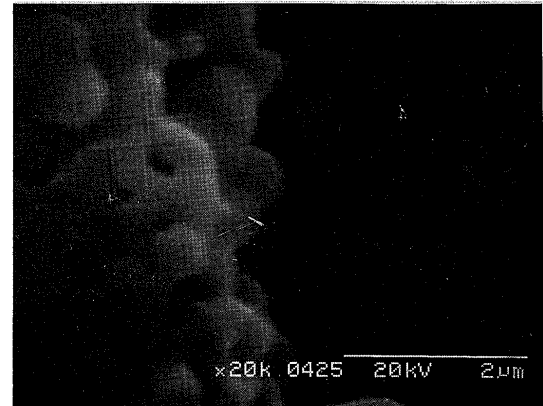
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Inner part

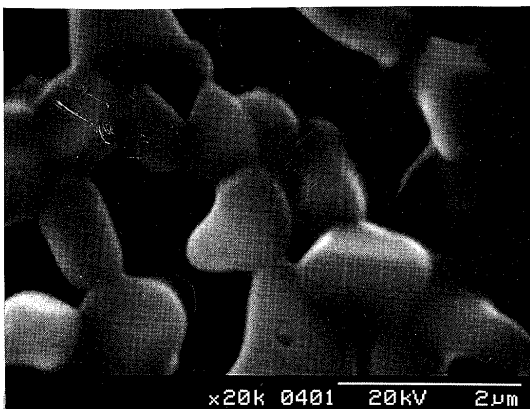


60 s, 90.2 % TD

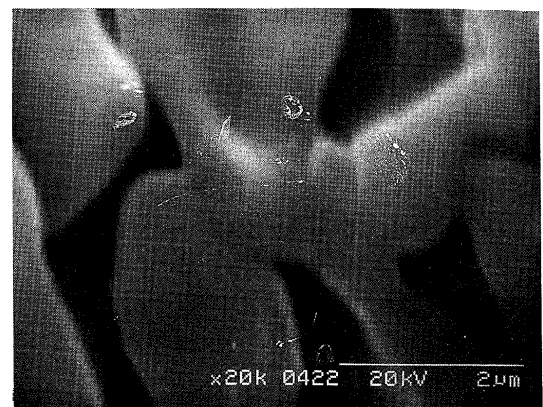


1200 s, 98.4 % TD

Surface



60 s, 90.4 % TD



1200 s, 98.4 % TD

Fig.5. Scanning electron micrographs showing changes in morphology in the inner part and on the outer surface exposed to plasma flow along green-to-fired transition.

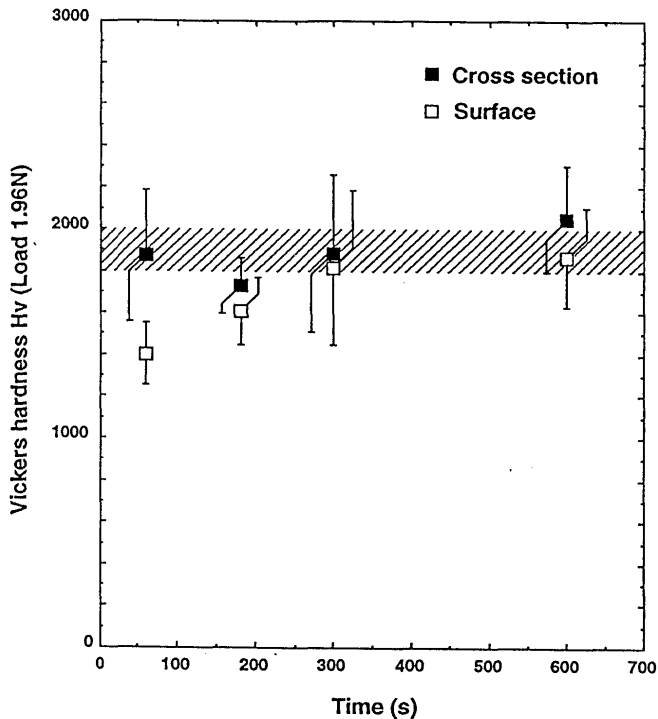


Fig. 6. Variation of Vickers hardness of pure Al_2O_3 samples as a function of sintering time..

part are plotted as open and solid squares, respectively, and the shaded portion in the figure correspond to the typical hardness data with various techniques. The highest achieved hardness exceeded 2000 Hv, which is relatively higher than those obtained with other techniques. At 60s the hardness on the surface reaches to a value of about 1400, while in the inner part it has a value of about 1800 and at about 300s the hardness for each position becomes similar to be about 1900. The reason for attaining smaller hardness on the outer surface than in the inner part may be attributed to the difference of the microstructures shown in Fig. 5; *i.e.*, larger grain size observed on the outer surface than in the inner part.

We have also examined the inclusion of nitrogen

into the samples by XRD and EPMA measurements and found nonexistence of any compounds of nitrogen.

4. Conclusion

Using a high power microwave N_2 plasmas at atmospheric pressure gas environment, ceramic sintering was performed for Al_2O_3 -based samples, by which rapid sintering was successfully obtained without any crack formation up to the final density higher than 95% TD. Remarkable difference in the grain growth during sintering was observed between the surface and the inner part, being inherent to the heat transport mechanism from the surface through heat conduction. The measured Vickers hardness was higher in the inner part than on the outer surface, corresponding to the microstructure of each position. The highest achieved hardness exceeded 2000 Hv.

Acknowledgment

The authors would like to express their gratitude to Dr. S. Sano for his critical comments on this work. They also thank to Mr. H. Ekou for his technical support.

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