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Densification and grain growth of alumina by microwave processing

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Abstract

Highly pure submicron alumina powdered compacts were sintered to a near theoretical density of 99.7% by 2.45 GHz microwave processing. Microwave heating showed enhanced densification processing and short sintering time as compared to conventional heating. However, a relatively large grain growth was observed at temperatures below 1400°C. It was found that there is a comparable grain size of 2.5 μm for the samples sintered by microwave and by conventional methods as they reached near theoretical density.

Keywords: Microwave sintering; Alumina; Densification; Grain growth

1. Introduction

There has been considerable interest in microwave sintering of oxide ceramics, especially for alumina materials. This is due to the fact that microwave heating offers the potential of enhanced sintering process and decreased densification temperature as well as shortened processing time for the materials [1–4]. In the 1980s, Meek et al. [5] reported that powdered compacts of 99.99% pure Al_2O_3 were sintered to a density of 91.7% using 2.45 GHz radiation. Janney and Kimrey [6] also observed that Al_2O_3 was densified far more rapidly by microwave heating than by conventional heating. The pellets of highly pure submicron Al_2O_3 powder were sintered to 97.76% of theoretical density at 1200°C/1.2 h in a single mode cylindrical cavity applicator CMPR-250 operating at 2.45 GHz in the TM_{012} mode [7]. De et al. [8] sintered the pure alumina green body to more than 99% theoretical density at 1500°C/30 min using a hybrid heating with microwave energy at 2.45 GHz. All the results mentioned above show that microwave sintering can accelerate the densification process and culminate in more uniform microstructures relative to conventional firing. On the other hand, however, large difference in grain growth by microwave processing has been reported. Some researchers [9] suggested that a smaller grain size can be obtained with microwave processing due to higher heating rates and shorter sintering cycle. For example, Patterson et al. [10] showed that grain sizes with 3.19 μm and 4.38

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μm were observed for the samples with 99.3% and 99.6% theoretical density by microwave and by conventional sintering, respectively. But Samuels and Brandon [11] found that the microwave-sintered samples have slightly larger grain sizes, as would be expected from their higher densities. From the different publications, it can be seen that the grain size strongly depends on the densification of alumina compact. In the present work, the densification and grain growth of highly pure submicron Al_2O_3 powder compacts were investigated by microwave sintering. The comparison between microwave and conventional heating in densification and grain growth of Al_2O_3 was also discussed.

2. Experimental

2.1. Sample preparation

A high purity (99.97%) alumina powder (Ceralox APA, Ceralox, Touscon, USA) with 0.05 wt.% of MgO as sintering aids, which has an average particle size of 0.4 μm and BET of 10 m^2/g, was used. The start powder was wet-ball milled with 3 mol% PVA and suitable amount of ethanol for 24 h to eliminate agglomerate. After drying and sieving, the powder containing binder was first uniaxially pressed at 100 MPa and then isostatically pressed at 250 MPa into the samples with $5 \times 6 \times 45$ mm^3. The binder of the green compacts was then burned out in a muffle furnace using a slow heating cycle, and resulting compacts had a green density of approximately 57%.

2.2. Sintering and measurement

The green samples were sintered in air using 2.45 GHz microwave energy. The microwave sintering system consisted of 0.78×10^{-2} m^3 multimode cavity and a continually adjustable power supply of 0.5–5 kW, which has been illustrated in detail elsewhere [12]. SiC rods (picket fence) susceptors, as described in Refs. [13,14], was used to initially hybrid heat low-loss alumina samples at relatively lower temperature. Microwave sintering was performed with a stable heating rate and a held of 30 min at peak temperature. The temperature of specimen in microwave oven was measured by a far inferred fiber optic pyrometer in the range of 550–2000°C, described in previous work [14]. For comparison, the samples were also sintered by conventional method with a fast heating rate of 200°C/h and a held of 2 h at designed temperature.

Bulk and relative density of the specimens in various final temperatures were obtained by the Archimedes method. The microstructure of the samples was observed by the OPTON-CSM 950 scanning electron microscope (SEM). The average grain size of the polished and thermally etched surface was determined by linear intercept method [15].

3. Results and discussion

3.1. Sintering and densification

Typical heating curve and power profile vs. time for microwave processing of the alumina are shown in Fig. 1. A constant power of about 0.5 kW was applied initially for half an hour, and then was gradually increased to the sintering temperature. After reaching critical temperature of near 800°C, the samples strongly coupled with electromagnetic field and resulted in considerable heating rate. It is very important to carefully control the heating rate to avoid thermal runaway during this period. Thermal runaway, which generally leads to thermal stress and damage of samples, was successfully eliminated by control of input power and use of hybrid heating structure in the present work. No cracks and deformation were observed in the samples after sintering.

Fig. 2 shows the variation of final density (expressed as a percentage of theoretic density) with

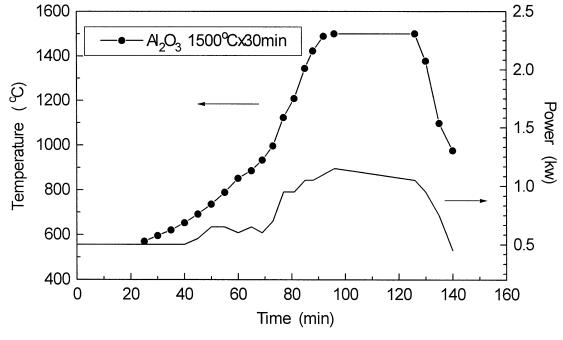

Fig. 1. Typical microwave firing schedule for alumina ceramics.

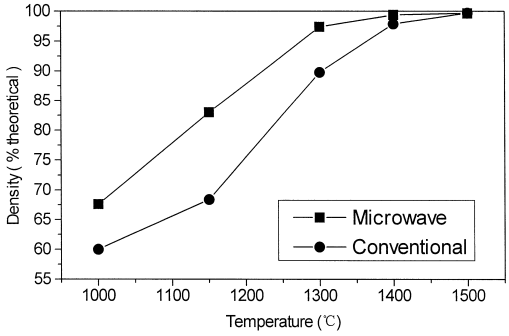

Fig. 2. Variation of final density with sintering temperature.

temperature for the sintered samples by microwave and conventional methods. It can be found that the microwave-sintered samples exhibit enhanced densification compared to their conventional-sintered samples at low sintering temperature (<1400°C), particularly at temperature below 1300°C. Microwave-sintered sample, for example, reached a 97.5% theoretical density at 1300°C with a held of 30 min, whereas conventionally-sintered sample reached the same density at 1400°C with a held of 2 h. A temperature difference of 100°C was observed between microwave and conventional processing for reaching 97.5% theoretical. However, samples were sintered to the same density of 99.7%, which is a near theoretical density, by microwave and conventional heating at the temperature of 1500°C. This implies that microwave heating can accelerate densification, but does not show higher density than conventional method for final density at 1500°C. Similar results can also be observed in Fig. 3 which illustrates density variation with holding time at 1500°C under two sintering methods. Although microwave-sintered sample reached 99.7% theoretical density in a shorter time of 15 min at 1500°C, conventionally-sintered sample also reached the same density with a longer holding time of 2 h. This final densification are consistent with results reported [10] where highly pure alumina powdered compacts were densified to 99.6% theoretical.

3.2. Grain growth and microstructure

SEM on fracture surface of samples by microwave and conventional sintering at 1300°C, 1400°C and 1500°C are given in Fig. 4. Fig. 5 shows the plot of average grain size vs. temperature. Clearly, microwave-sintered sample displayed larger grain size than that in conventional-sintered sample at the same temperature. This is consistent with their higher density. For example, the grain sizes are 1.2 and 0.7 for microwave and conventional sintering at 1300°C, which corresponds to their densities of 97.5% and 89% theoretical, respectively. These results suggest a rapid grain growth and was also accompanied by the enhanced densification in microwave sintering of ceramics at low temperature. Previous work [14] on Ce–Y–ZTA composites also revealed that alumina grain sizes of microwave-sintered samples are larger than that of conventional at temperature below 1550°C.

In general, grain growth law during isothermal holding is described as:

$$G^n - G_0^n \propto (D/T)t \tag{1}$$

$$D = D_0 \exp(-\Delta E/RT) \tag{2}$$

where G and G_0 are the grain sizes at holding time $t = t$ and $t = 0$, n is a constant related to grain growth mechanism, D is diffusion coefficient related to the grain, and ΔE is diffusion activation energy. The equations are thought to be applicable for the solid state sintering of Al_2O_3. Janney and Kimrey [6] calculated and found that the apparent activation energy for sintering of alumina is 160 and 575 kJ/mol under microwave and conventional methods, respectively. Low activation energy and increased diffusion coefficient would be responsible for the fast grain growth in microwave processing.

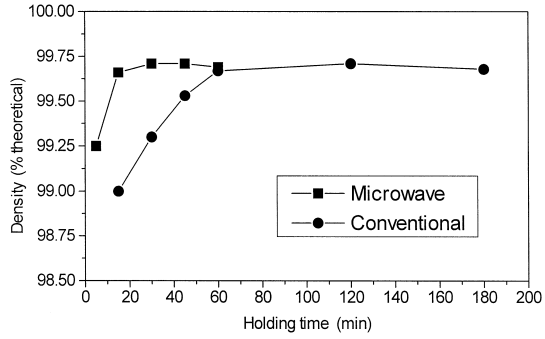

Fig. 3. Variation of final density with holding time at 1500°C.

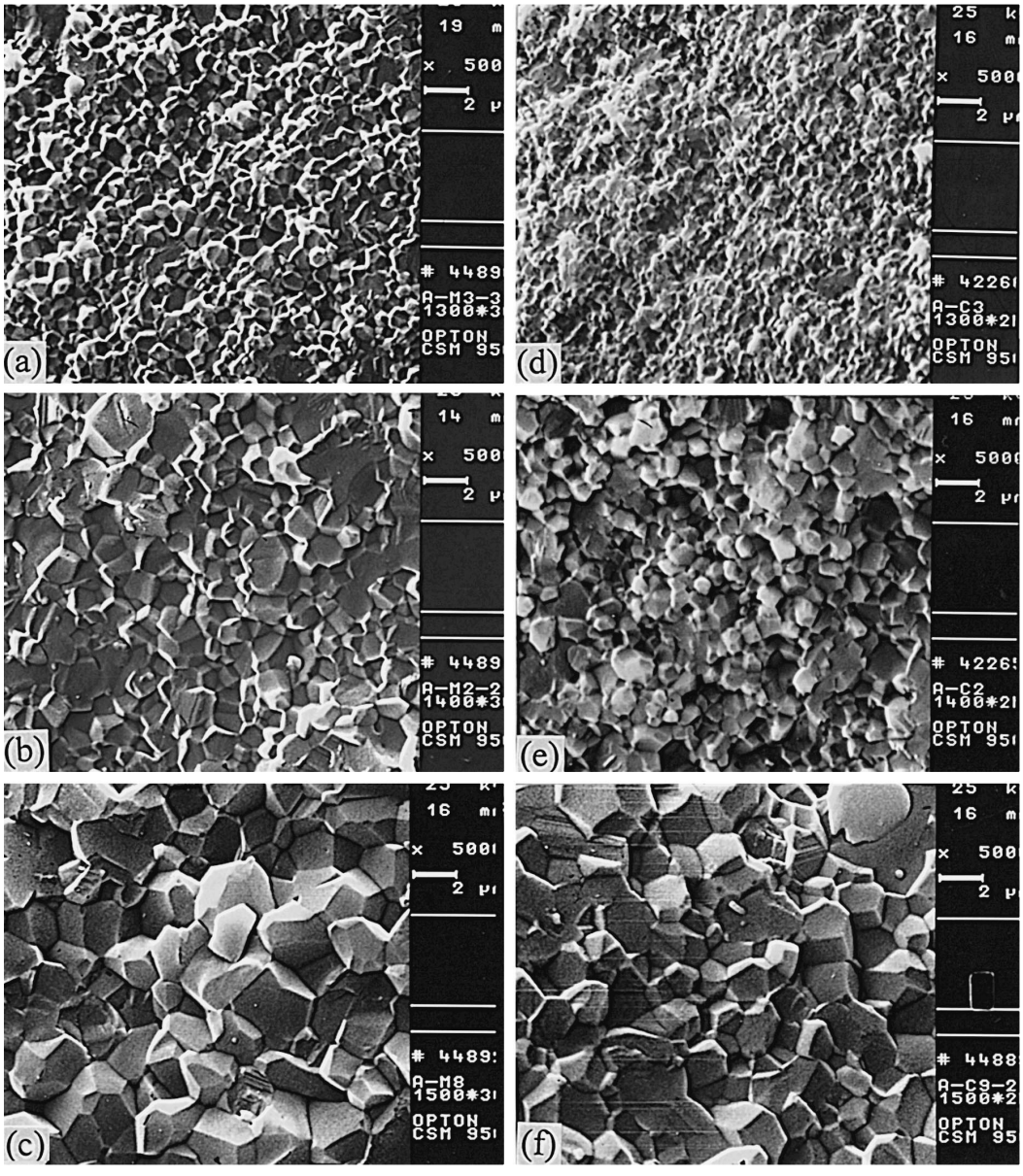

Fig. 4. SEM micrographs of samples sintered in microwave furnace at (a) 1300°C; (b) 1400°C; (c) 1500°C; and in conventional furnace at (d) 1300°C; (e) 1400°C; (f) 1500°C.

Because the grain size strongly depends on the density of sintered samples, the comparison of grain growth between the two methods should be determined at the same density. Fig. 6 shows the variation of grain growth with density for microwave and conventional sintering, where density varies from about 60% to near theoretical density of 99.7%. It can be found that there is a comparable grain size at the same densification for the two methods. In addition, grain growth with densification has the same

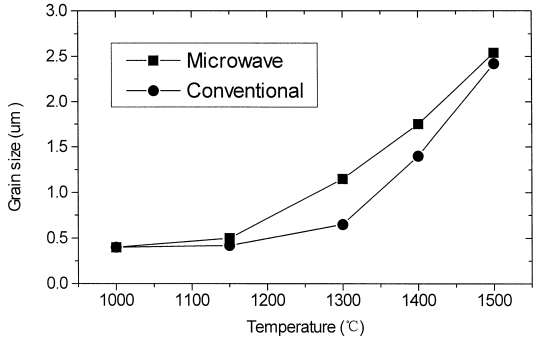

Fig. 5. Grain sizes vs. temperature for two sintering methods.

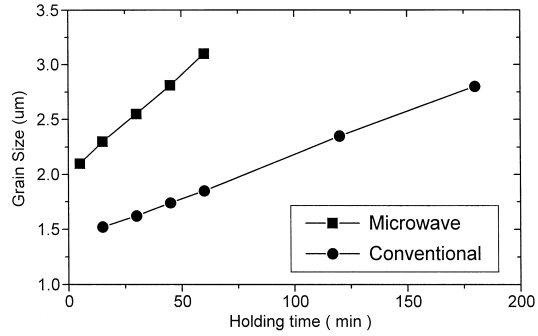

Fig. 7. Influence of holding time on grain growth at final temperature of 1500°C.

increasing tendency, and a significantly grain growth was observed from 95% to more than 99.5% theoretical density.

Grain growth in ceramics is a function of both the sintering temperature and holding time at this temperature. Therefore, the grain size in sintered samples also relied on the holding time at final sintering temperature. Fig. 7 illustrates the influence of holding time on grain growth at final temperature of 1500°C. Microwave-sintered samples show a largely grain growth rate compared to the conventional. It should be noted that after reaching near theoretical density of 99.7% with a hold of 15 min and 2 h for microwave and conventional, the density of samples can not be increased with longer holding time and excessive grain growth. The reason why the results show a more grain growth vs. holding time in microwave processing is not clear. Perhaps the increased coupling with microwave fields and diffusion of mass transport at final temperature was responsible for the rapid grain growth. Moreno et al. [16] also showed that grain size larger than 4 μm was observed by microwave sintering with a held of 1.5–50 h at 1550°C for highly pure alumina with MgO dopant. In addition, Janney et al. [17] had exhibited that the kinetics of grain growth were greatly increased by the 28 GHz microwave anneals, e.g., the grain growth rate at 1500°C in the microwave furnace was the same as the rate at 1700°C in the conventional furnace.

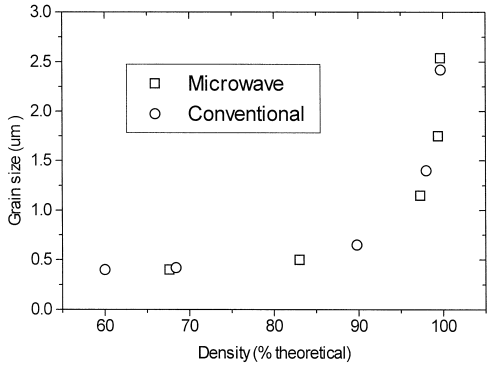

Fig. 6. Grain sizes vs. density for two sintering methods.

4. Summary

A comparative study of microwave and conventional sintering on densification and grain growth of highly pure submicron alumina powdered compacts has shown that the samples can reach a near theoretical density of 99.7%, and there is a comparable grain size of 2.5 μm. Although the densification processing was enhanced and sintering of samples was finished with shorter time for microwave heating, the increased grain growth with fast densification was also observed. Therefore, microwave-sintered samples tend to have larger grain size at the temperature below 1400°C due to their enhanced densification. However, there is almost the same grain size in samples sintered by microwave and by conventional as they have identical densification. In addition, microwave-sintered samples show a largely grain growth rate with holding time at final temperature of 1500°C compared to conventional after reaching near theoretical density of 99.7%.

Acknowledgements

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