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MICROWAVE SINTERING OF LARGE ALUMINA BODIES

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ABSTRACT

The application of microwaves as an energy source for materials processing of large alumina bodies at elevated temperatures has been very limited to date. Due to the restrictions inherent to the process, most work has concerned itself with small laboratory samples. The nonuniformity of the microwave field within a cavity subjects large alumina bodies to areas of concentrated energy, resulting in uneven heating and subsequent cracking. Smaller bodies are not significantly affected by field nonuniformity due to their smaller mass. This work will demonstrate a method for microwave sintering of large alumina bodies while maintaining their structural integrity. Several alumina configurations were successfully sintered using a method which creates an artificial field or environment within the microwave cavity.

INTRODUCTION

Microwave sintering of large alumina bodies has proven difficult due to the restrictions imposed by the microwave field. A multimode cavity operating at 2.45 GHz contains an inherently nonuniform electromagnetic field. Small laboratory samples can be sintered in this field with relative success. Multiple laboratory samples have also been simultaneously sintered using the proper insulation configuration[1]. Multiple sintering represents an increase in total mass, but is not a significant increase in the volume of the individual parts. Although successful, these processes and insulation configurations were not applicable to larger alumina bodies. As the work piece increases in mass and cross section it becomes subject to cracking and warping, due to the nonuniform field, which creates hot spots or areas of concentrated energy. Coupled with this condition is the necessity of selecting insulation materials which are both transparent to the microwave field and able to withstand elevated temperatures. To operate in such a hostile environment it was necessary to create an artificial environment within the microwave cavity. This work will demonstrate the fabrication of such an environment in which large alumina bodies were sintered crack free to high densities.

THEORY OF MICROWAVE HEATING

The power dissipated by a ceramic in an electric field creates heating. The power dissipated is given by the following equation[2].

$$P = (2\pi f \epsilon')(E^2/2) \tan \delta \qquad (1)$$

where P is the power dissipated, f is the frequency, ϵ' is the dielectric constant, E is the electric field strength, and $\tan \delta$ is the loss tangent for the dielectric. This illustrates that the ceramic must have an appreciable loss tangent and dielectric constant in order for significant heating to occur. The depth of microwave penetration into the sample is quantified by the skin depth which is the distance at which the electric field falls to 1/e (which is equal to 37%) of the field strength at the surface. The skin depth is calculated from the following formula[3]:

$$\text{Skin depth} = 1/(\pi f \mu \sigma)^{\frac{1}{2}} \qquad (2)$$

where μ is the magnetic permittivity and σ is the dc conductivity. High-purity alumina, such as that used in this study, is largely transparent to 2.45 GHz microwaves at room temperature; the skin depth is of the order of meters.

MICROWAVE FACILITY

All experimentation was conducted in a 0.6096 m^3 (2 ft^3) resonant, microwave cavity operating at a frequency of 2.45 GHz with a maximum power level of 6 kW. The cavity, although not airtight was operated with a flowing argon atmosphere. The temperature was monitored using an Accufiber[a] fiber optic thermometer which passed through the cavity wall and was sighted next to the work piece. A slug tuner mounted in the cavity was used to reduce reflected microwave power and to enhance microwave coupling to the work piece.

MATERIALS AND SAMPLE PREPARATION

Three configurations of alumina bodies were used in this study. The first were alumina disks 0.157 m x 0.0127 m (6.1875" x 0.5")

[a] Accufiber, Inc., Vancouver, WA.

and 0.157 m x 0.019 m (6.1875" x 0.750") which weighed 545 grams and 925 grams respectively. These disks were cold pressed from AKP-50[§] powder which has a mean particle size of 0.20 μm and a purity of 99.99%. The green and sintered disks were 55% and 93% of theoretical density respectively. The second shape was a commercially supplied hexagonal tile of AD995[†] alumina 0.1397 m (5.50") point to point x 0.0127 m (0.50") weighing 420 grams and containing a binder. The green and sintered hexagonal tiles are compared in Fig. 1, the degree of densification, 96% of theoretical density, is obvious by comparing the two tiles. The third shape was also a commercially supplied tile of AD-90[†] alumina 0.1016 m x 0.1016 x 0.0127 m (4" x 4" x 1/2"), weighing 525 grams and containing a binder. The AKP 50 disks were formed by uniaxial pressing at 170 psi. without binders or sintering aids. The commercial preformed hexagonal and square tiles were preheated to 800°C for 10 hours to remove the binder prior to microwave sintering.

SINTERING PROCEDURES

The alumina work piece was housed within a double enclosure of carbon and alumina insulation. An inner box of carbon immediately surrounded the sample followed by an outer box of low density alumina insulation. Fig. 2, shows the sample with the top carbon disk and alumina insulation removed, resting in the low density alumina insulation box. Temperature was monitored by insertion of an optical fiber thermometer. The assembled configuration was slowly heated in an argon atmosphere from room temperature to 1600°C with an isothermal hold of 10 minutes. The total processing cycle required 7-8 hours. To avoid thermal cracking the samples were slowly cooled by reducing the power until reaching 1300°C at which time the power was turned off. Slow cooling continued with the alumina insulation containing the heat until reaching room temperature.

DESIGN OF INSULATION

A low density commercial alumina insulation board[Δ] (480.54 kg/m^3) containing 20% SiO$_2$ was used to contain the work pieces during sintering. The insulation served several purposes, one is to contain the heat during sintering, another is to prevent arcing within the microwave cavity. Initial attempts at sintering the alumina work pieces housed only within the alumina insulation met

[§] Sumitomo Chemical Co., Ltd., Osaka, Japan

[†] Coors Ceramics Co., Golden, CO.

[Δ] Zircar Products Inc., Florida, NY

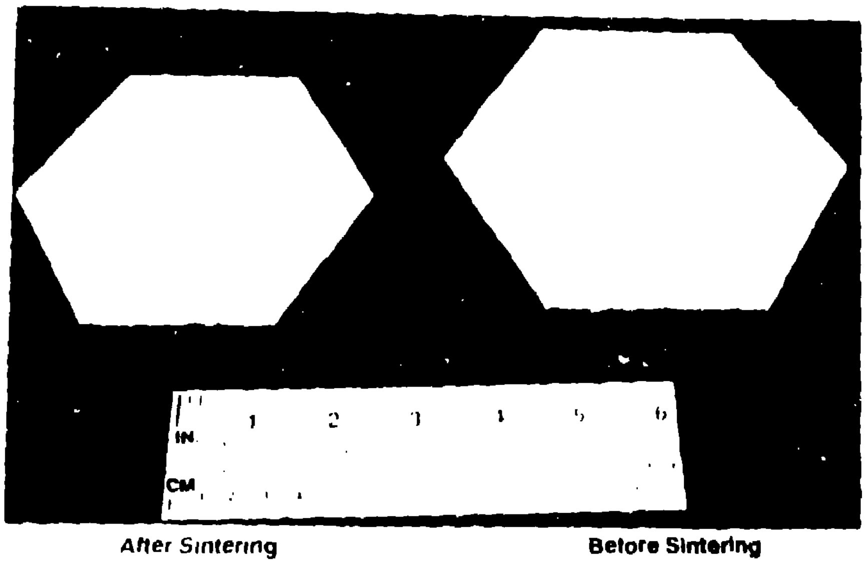

Figure 1. Green and microwave sintered hexagonal tiles of Coors AD995 alumina. The microwave sintered tiles had an average density of 93% and weighed 420 grams.

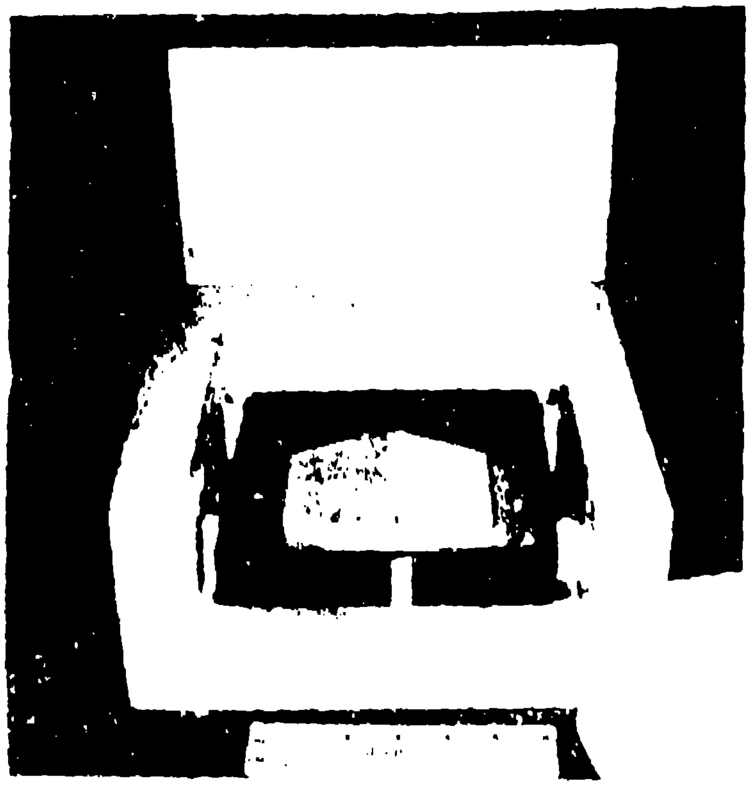

Figure 2. Samples were sintered in an inner box of carbon which was then surrounded by an outer box of low density alumina insulation. The top alumina insulation board and carbon disk were removed for this picture.

with poor results. Unlike small alumina samples the large alumina bodies cracked or completely shattered during sintering. This resulted from an increased mass of working material within a nonuniform microwave field. Methods to create a uniform field within the cavity such as using a stirring fan or a rotating table have only met with limited success. It was therefore necessary to create a system in which the alumina work pieces were surrounded by a uniform microwave field while contained within a nonuniform cavity. A system which would also allow the work piece to rise slowly to its sintering temperature and eliminate thermal cracking.

ARTIFICIAL MICROWAVE FIELD

Two insulation materials were selected to contain the alumina work pieces. A low density alumina insulation board was selected for its transparency to microwaves and its ability to withstand relatively high temperatures. The second material, a low density carbon was used to surround the work piece. The carbon was selected because of its proven ability as a setter material for alumina, and it also acts as a microwave susceptor, transferring heat uniformly. For heat containment in the microwave cavity a box was fabricated 0.2794 m (11") square x 0.0889 m (3 1/2") high x 0.0381 m (1 1/2") thick of the low density alumina insulation. Inside this insulation box a carbon assemblage was framed around the alumina work piece. Within the alumina insulation box, a 0.00635 m (0.250") thick bed of 60 mesh alumina grain was evenly spread on the bottom board of insulation. Upon this bed rested a carbon disk 0.168275 m (6.625") x (0.125") 0.003175 m which in turn held the alumina work piece. A thin layer of alumina grain was spread between the carbon disk and work piece to insure ease of movement as the alumina shrunk during sintering. This also prevented any cross contamination of the materials. Two additional carbon disks of the same size were placed on top of the work piece, again with alumina grain between the carbon and the work piece. The edges of the alumina work piece were boxed in on four sides with two carbon strips per side, each measuring 0.17145 m (6.750") x 0.03175 m (1.250") x 0.003175 m (0.125"). These strips rested on edge supported by the top carbon disks, completely enclosing the work piece. A hole was drilled through one wall of the insulation and through two carbon strips to allow passage of a optic fiber light pipe to monitor temperature. This assembled configuration was slowly heated in a flowing argon atmosphere to prevent carbon oxidation at the elevated temperatures. The heat cycle of 7 to 8 hours consisted of a slow rise to 1600°C and an isothermal hold for 10 minutes followed by a slow cool down to room temperature (See Fig. 3), reducing thermal shock. Although these procedures proved successful, some cracking occurred in a small percentage of the samples. This was attributed to incomplete binder bake out of the hexagonal tiles. The presence of binders in the samples during microwave sintering resulted in selective coupling to the binder and subsequent uncontrolled rapid heating and cracking of the sample. Also contributing to the cracking was degradation of the carbon enclosure, due to an inadequate argon atmosphere in the loosely sealed cavity. The problems were easily resolved by increasing the bake-out time for binder removal, and additional argon flow during microwave sintering.

POWER USAGE

Typical input and reflected power curves versus time for an AD995 hexagonal tile are shown in Fig. 4. The reflected power is almost negligible compared to the input power. Reflected power was not even measurable until reaching 2700 W of input power, at which time it only amounted to 1% of the total power. The peak reflected power of 8% was reached at a total input power of 48%. As the reflected power is not recoverable, the total power usage was 48% of the available power of 6 kW. This amounted to a peak power usage of 2880 W to sinter an alumina hex tile. This represents a more efficient use of energy as compared to microwave sintering of much smaller laboratory samples[1]. The scale-up in mass lowers power consumption considerably, an important aspect as applied to industrial processing. If processing time or time at peak temperature is considered, an additional savings in energy is realized. Alumina samples weighing as much as 925 grams were microwave sintered within 7 to 8 hours, as compared to conventional sintering which may require 24-48 hours for the sintering and cooling process. It is only practical to compare energy costs for microwave sintering with conventional electrical sintering, as natural gas is a cheaper energy source[4]. Again, the relatively short cycle time for microwave processing may weigh well against the cost differences with natural gas.

RESULTS

Although the microwave field within the cavity remained nonuniform, the carbon enclosure created an artificial microwave field surrounding the alumina work piece. This allowed for slow, uniform heating producing dense, crack free alumina bodies. The alumina work piece would suscept itself at elevated temperatures, however, it was necessary to house it in the carbon enclosure for even heat distribution and to control the heating cycle. Sintering appeared uniform throughout the samples, no warpage was evident and dimensional tolerances were maintained. The theoretical densities were 92-94%. Ceramics produced by this technique

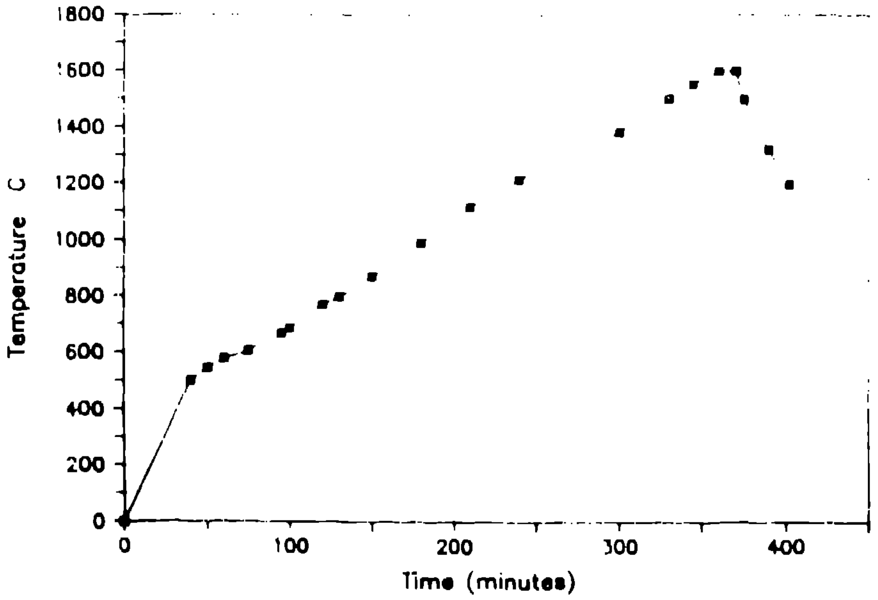

Figure 3. The time-temperature cycle was a total of 7 to 8 hours. It consisted of a slow rise to 1600°C and an isothermal hold for 10 minutes followed by a slow cool down to room temperature.

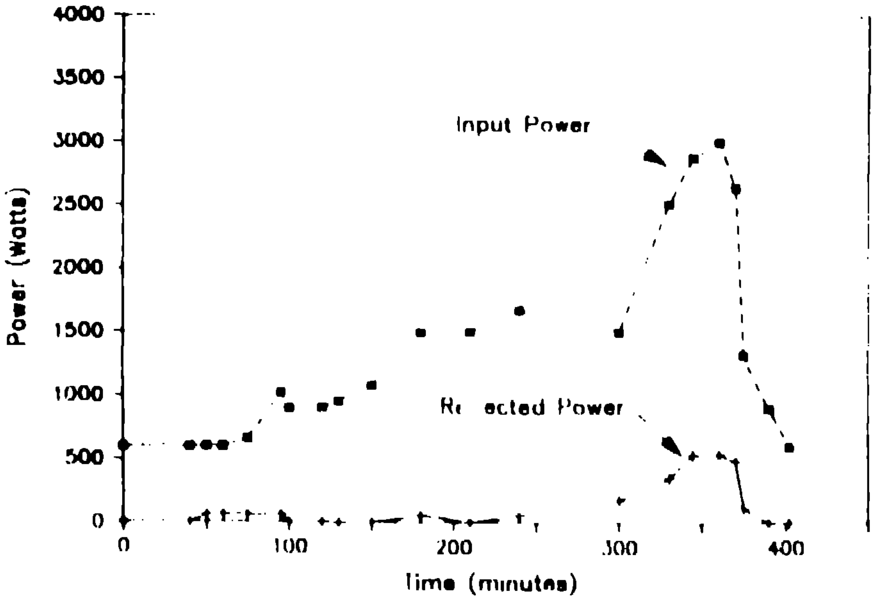

Figure 4. The reflected power is almost negligible compared to the input power. Reflected power was not even measurable until reaching 2700 W of input power, at which time it only amounted to 1% of total power.

are of good quality. Densities are comparable within 4 or 5% to those which result from conventional processing. The average power level needed for sintering the alumina tiles was 45% of the maximum level of 6 kW or 2.7 kW. The sintering cycle required 7 to 8 hours from room temperature to cool down. Since the completion of this work, additional like samples have been successfully sintered within a 5 hour cycle. These alumina samples represent a vast increase in volume over previously sintered parts and are believed to be the largest high purity alumina bodies sintered by microwave processing. Also of significance is the relatively short processing time and the variety of shapes involved. The results have shown that microwave sintering of alumina can be achieved beyond small laboratory samples and may be a viable alterative to conventional processing methods.

FUTURE WORK

This work has described a successful method for microwave sintering of large alumina bodies, but more work needs to be done to optimize the processes to accommodate production operations. Toward that end a new pressure/vacuum microwave cavity is being put on line which will allow a tight atmosphere, and may produce a more uniform microwave field due to its cylindrical shape. For production purposes the alumina insulation and carbon susceptor need to become an integral part of the microwave cavity and be capable of withstanding repeated usage. With these and perhaps other necessary refinements in place, full scale production is possible.

REFERENCES

1. J.D.Katz and R.D.Blake, Ceramic Bulletin, Vol. 70, No. 8, pp. 1304-1308, 1991.

2. "*Introduction to Ceramics*", W.D.Kingery, H.K.Bowen and D.R.Uhlmann, John Wiley and Sons, New York, 1976, p. 927.

3. "*Dielectrics and Waves*", A. R. Von Hippel, John Wiley and Sons, New York, 1954, p. 61.

4. S.Das and T.R.Curlee, Am. Ceram. Soc. Bull., 66, pp. 1093 1094, 1987.