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MICROWAVE (HYBRID) HEATING OF ALUMINA AT 2.45 GHZ: I. MICROSTRUCTURAL UNIFORMITY AND HOMOGENEITY

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Microwave (hybrid) heating (MHH) is a unique combination of microwave (MW)-material interaction and conventional radiant/conduction mechanisms that facilitates the attainment of very high heating rates in a 2.45 GHz, multimode MW cavity. Microstructural uniformity and homogeneity of dry-pressed green samples of pure, undoped alumina with (MHH) relative to conventional fast firing (CFF) has been studied. The confluence of the two heating mechanisms (with MHH) results in an improved parity in temperatures across specimen cross-sections vis-a-vis CFF and stand-alone MW (SMW) sintering. This enhanced parity in temperatures (with MHH) can be said to be responsible for the better microstructural homogeneity and improved mechanical properties relative to CFF. Sintering of larger (20 gm vs. 6 gm) samples with MHH shows evidence of a definitive mass dependance on the MHH phenomena. Larger masses show a better parity in temperatures between the surface and interior of the sample. Consequently, this results in enhancements in the homogeneity of the microstructure, and improved and more uniform mechanical properties relative to the smaller MHH and CFF samples.

INTRODUCTION

MW sintering is a technique that offers enormous potential for the fabrication of ceramics and ceramic composites with improved microstructures. Work in this area by a number of researchers [1-7] demonstrate the efficacy of MW sintering from the standpoint of lower sintering temperatures and smaller grain sizes compared to conventional sintering. This has been attributed to enhanced diffusion and a lower activation energy for sintering that is characteristic of MW energy [2-4].

In order to apply the benefits of ultra rapid heating to MW sintering, a technique that makes use of both MW-material interaction as well as conventional radiant/ conduction heating mechanisms has been developed. 'Hybrid heating using MW energy' (MHH) [8-10] makes use of radiant/conduction heating to rapidly heat samples through low temperature regimes to a critical temperature above which the microwaves couple readily with the material. A heating rate of up to 750°C per minute for pure, undoped alumina in a 2.45 GHz, 6.4 KW (max.) multimode cavity, making use of less than half the peak power output, has been attained with this technique.

Microstructural homogeneity (parity in grain sizes and porosity across sample cross-sections) is difficult to achieve with conventional techniques. Although stand-alone MW (SMW) sintering has been surmised to result in more homogeneous microstructures compared to conventional techniques [2], no evidence to support that premise has been presented. Past efforts (with SMW sintering) [7] indicate non-uniformities in the form of appreciable differences in grain sizes between the surface and interior of sample cross-sections.

It is suggested that an improved parity in temperatures between the surface and the interior, that is unique to MHH is responsible for the enhancements in microstructural homogeneity and mechanical properties relative to CFF.

EXPERIMENTAL PROCEDURE

A pure, undoped, commercially available alumina was used. The alumina designated A-16* was a 99.97% pure alumina grade (as received) with an average particle size of 0.48 μm, a broad particle size distribution, and a surface area of 4.697 m^2/gm.

The experimental set-up for the MHH is illustrated in Figure 1. MHH was effected in static air in a 2.45 GHz, 6.4 KW (max) ** multimode cavity, with the sample enclosed in a silicon carbide lined susceptor. Temperature monitoring was effected by inconel shielded 'K'-type thermocouples#(for temperatures up to 1100°C) and by two color infrared pyrometry## (for temperatures above 1100°C). A peak heating rate of the order of 1500°C in 120 sec (750°C per minute) for pure undoped A-16 alumina at a power output of around 3 KW was attained.

CFF (conventional fast firing/ isothermal sintering) was effected in static air by inserting the samples into a resistance heated tube furnace+ which was preheated to the sintering temperature. The samples were stationed in alumina boats which in turn, were slowly inserted into the heat zone of the tube furnace. Temperatures were read off a 'B' type Pt-Rh thermocouple #(with digital display) which was kept in contact with the alumina boat and the walls of the alumina tube of the furnace.

In order to understand the heating mechanisms (and temperature gradients across the sample cross-sections) characteristic of MHH and CFF, experiments designed to obtain simultaneous surface and interior temperature profiles under actual conditions of heating were initiated. The experiments comprised stationing inconel shielded 'K' type thermocouples on the surface and in the interior of the samples, and heating the samples as shown in Figure 2. The samples were heated under conditions of SMW heating, MHH and CFF. Twenty five gm dry-pressed green samples, with a cavity (core) at the interior were used. For the MHH experiments, in addition to the 25 gm sample, an 8 gm dry-pressed sample was also used (to study the effect of mass on the MHH). For the CFF, the specimen had to be pre-sintered to around 1000°C., for sufficient green strength, and to be able to withstand the high heating rates and thermal stresses.

In order to compare ultra rapid sintering using MHH with CFF, and study the mass dependence of the two modes of heating, 6 gm and 20 gm cylindrical pellets of A-16 alumina were cold pressed to 4000 psi. (green density~52%). These were then sintered in the MW oven under hybrid heating conditions at 1500°C, with a holding time of 30 minutes. Identical green compacts were also fired in the preheated resistance tube furnace under isothermal heating conditions at the same temperatures and holding times as the MHH samples.The samples were then scanned from the surface to the interior as shown in Figure3. The five positions from the surface to the interior and back to the surface have been designated A, B, C, D, and E, respectively.

Bulk and relative densities of the sintered samples were obtained by the Archimedes density method. The samples were then polished using standard ceramographic techniques, and thermally etched at a temperature of 1450°C for 30 min. Microstructures representative of both the surface and the interior of each sample were obtained by a scan of the transverse cross-section of the polished and etched sample section in the SEM, as shown in Figure 3. Grain intercept lengths and porosities were

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* Aluminum Company of America (Alcoa) Inc., Pittsburgh, PA.
** Model Radarline QMP 2101B-6, The Raytheon Company, Waltham, MA.
# Omega Engineering Inc., Stamford, CT.
Ratio Scope 8, Capinter Inc., Fair Lawn, N.J.
+ Model Sola-Basic, Lindberg, Watertown, WI.

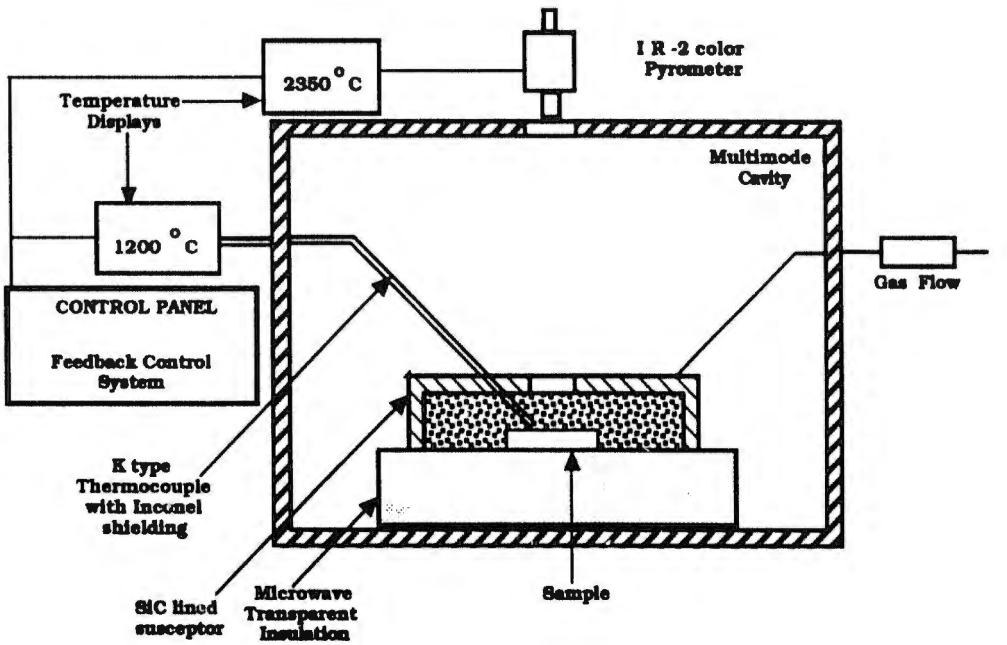

Figure 1. Schematic of multimode 2.45 GHz., 6.4 KW (max.) MW cavity.

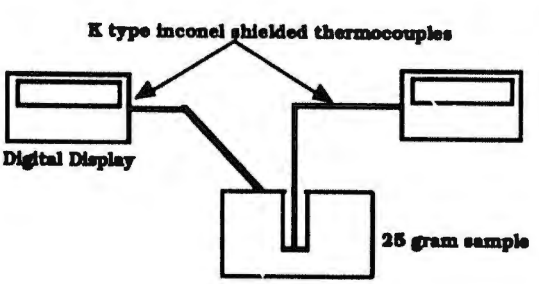

Figure 2.. Arrangement used for surface-interior temperature profile measurements.

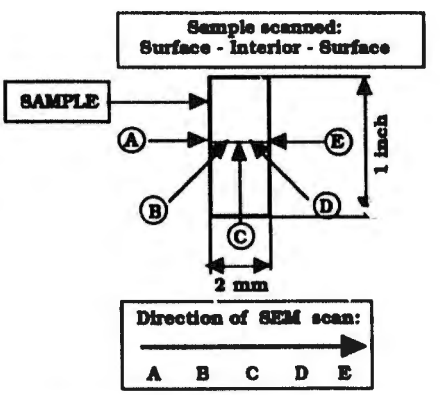

Figure 3. Sample scan in the SEM

computed from the SEM micrographs using standard quantitative stereological techniques. Average grain sizes were determined from measured average lineal grain intercept lengths according to the equation [11]

$$\text{grain size} = 1.56 \bullet \text{grain intercept length} \quad (1)$$

The resulting data were plotted, correlated and compared .

RESULTS AND DISCUSSION

Surface-Interior Temperature Profiles

Simultaneous surface-interior temperature profile measurements were carried out for samples heated by stand-alone MW heating (SMW), conventional fast firing (CFF), and MW (hybrid) heating (MHH). A uniform temperature distribution across the cross-section is vital to densification (and coarsening), and is perhaps, the single most important factor contributing to microstructural homogeneity. Twenty five gm dry pressed samples for all three techniques were employed. In addition to this, the temperature profile for an 8 gm sample heated by MHH was also obtained.

Figure 4(a) is an illustration of the temperature profile obtained for the pre-sintered 25 gm sample, heated to 1200°C by CFF. As is evident from the profiles, for the short soaking times (~ 30 minutes) employed for sintering the CFF samples, the interior of the samples, for most of the sintering time, do not experience the same thermal histories as the sample surfaces. These differences in temperature at the surface and the interior translate into microstructural inhomogeneities, as will be discussed in the following sections.

Figure 4(b) shows the surface interior temperature profile obtained for the alumina sample subjected to SMW heating. The alumina did not heat up beyond 500°C, even with power levels as high as 5 KW. However, even at the low temperatures that the sample was heated to, the interior of the sample was at a significantly higher temperature than the surface. This is a direct consequence of the inverse temperature gradients that is characteristic of SMW heating. This disparity in temperatures between the surface and the interior may be expected to increase with an increase in temperature.

Figure 4(c) is a plot of the temperatures experienced by the surface and the interior of an 8 gm sample heated by MHH.When the MW power is switched on, the SiC from the susceptor heats up quickly. This in turn, transmits heat to the surface of the alumina sample by radiation. The surface of the alumina specimen heats up and conducts heat to its interior by phonon conduction. This process continues until the entire sample heats up to a temperature of about 800°C, above which, the alumina absorbs microwaves readily. The onset of MW heating causes the interior of the sample to heat up (owing to the inverse thermal gradients characteristic of MW heating). However, the surface of the sample which receives heat from the susceptor, also conducts heat into the interior of the sample. Thus, the interior of the sample which is subjected to heat from these twin sources, heats up to a higher temperature than the surface. Besides, heat is also lost from the sample surface by dissipation, to the atmosphere, as well as the refractory bricks below. This disparity in temperatures (40-60°C at 1200°C) between the surface and the interior is reflected in Figure 4(c).

The surface-interior temperature profile for the 25 gm sample heated by MHH is illustrated in Figure 4(d).The heating process is very similar to that for the 8 gm hybrid heated sample. However, owing to the larger mass the process of heat conduction to the interior takes a longer time. Equivalent temperatures are realized at the surface and interior of the specimen throughout the sintering (soaking) time.

It is this parity in temperatures that is responsible for the more uniform and homogeneous microstructures obtained with the MHH process relative to CFF. Since the masses of the specimens used for sintering studies (6 gm and 20 gm) are very close to

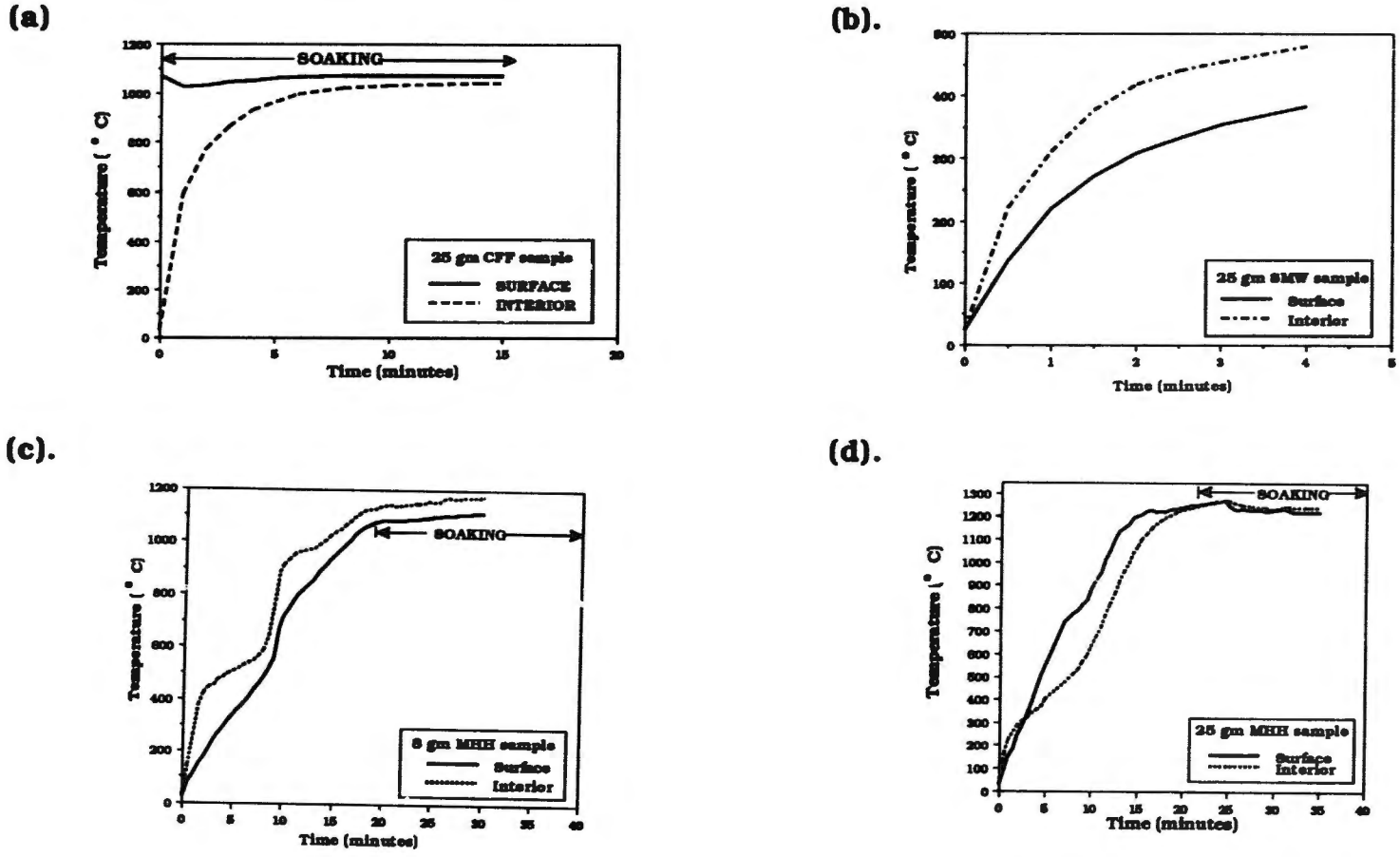

Figure 4. Surface-interior temperature profiles of: (a). 25 gm (pre-sintered) CFF sample; (b). 25 gm SMW sample; (c) 8 gm MHH sample and (d) 25 gm MHH sample.

those employed for the surface-interior temperature profiles (8 gm and 25 gm), the profiles were used to explain the differences in the sintering phenomenologies, and the consequent disparities in microstructures.

MW (Hybrid) Heating vs. Conventional Fast Firing of A-16 Alumina : Mass Dependence

In order to effect a comparison of MHH and CFF, identical 6 gm dry-pressed samples of A-16 alumina were sintered using both techniques at 1500°C, with a holding time of 30 minutes. Previous work by Dé et al. [9-11] have elucidated these results in detail. The surface-interior temperature profiles showed that a better volumetric heating and a significantly better parity in temperatures was observed with the larger samples. Subsequent efforts were therefore directed towards sintering larger (20 gm) dry-pressed samples under the same state conditions as the smaller samples (1500°C for 30 minutes) by both MHH and CFF. The 20 gm. CFF samples did not survive the excessive thermal stresses caused by the temperature gradients (thermal shock) and fractured into fragments each time.

Representative transverse cross-sections of sample portions (polished and thermally etched) were then scanned from the surface to the interior as shown in Figure 3. The results from the microstructural analysis and stereological computations for the 20 gm MHH specimen were incorporated with those from the smaller (6 gm) MHH and CFF samples, and will be discussed here.

The relative densities of the 6 gm MHH and CFF samples were 96% and 87% (of the theoretical), respectively, while that of the 20 gm. MHH sample was 98% of the theoretical, sintering having been carried out under the same state conditions of time and temperature.

Figure 5(a) is a plot of the volume fraction of porosity (computed stereologically) as a function of the sample thickness. The 6 gm CFF sample showed very significant differences in porosity from the surface to the interior with a maximum porosity of around 23% at the center, whereas the porosity for the 6 gm MHH sample ranged from 4.06 to 5.8%. The porosity for the 20 gm MHH sample was only 2%.

Figure 5(b) is a plot of the ave. grain intercept size (GI) versus sample thickness for the 3 samples. The 6 gm MHH sample exhibited somewhat larger GI sizes than the CFF sample (average GI size -1.41 μm relative to 0.70 μm for the CFF sample), with the largest grains at the center. The CFF sample (6 gm) showed an appreciable variation in the GI size distribution across the cross-section, with grains progressively increasing from surface towards the interior, and then dipping drastically in size at the interior of the specimen. The ave. GI size of the 20 gm MHH sample was 0.75 μm, with very little variance across the sample cross-section.

The enhanced microstructural uniformity and homogeneity for the bulk MHH sample (in spite of the inhomogeneities in the starting green microstructure) can be explained by the temperature distribution across the sample cross-section. As discussed earlier (Figure 4 (d)), the bulk (20 gm) MHH sample experiences nearly the same temperature (1500°C) across it's entire cross-section for the full length of the soaking time due to the confluence of the twin heating mechanisms that characterize the MHH process. This is in contrast to the temperature gradients experienced by the 6 gm CFF sample (interior significantly cooler than the surface which is at the sintering temperature) and the 6 gm MHH sample (sample interior at an appreciably higher temperature than the surface which is at the sintering temperature). It is this equanimity in temperatures between the surface and the interior of the bulk MHH sample for the entire period of sintering, that culminates in the enhanced densification (microwave enhanced diffusion) relative to the CFF specimen, and reduced grain growth as compared with the 6 gm MHH sample. Further, it is this parity in temperatures that is responsible for the dramatically improved microstructural uniformity throughout the bulk (20 gm) MHH specimen.

The microstructure-mechanical property correlation is reflected in plots of the mechanical properties (microhardness, fracture toughness) as a function of the

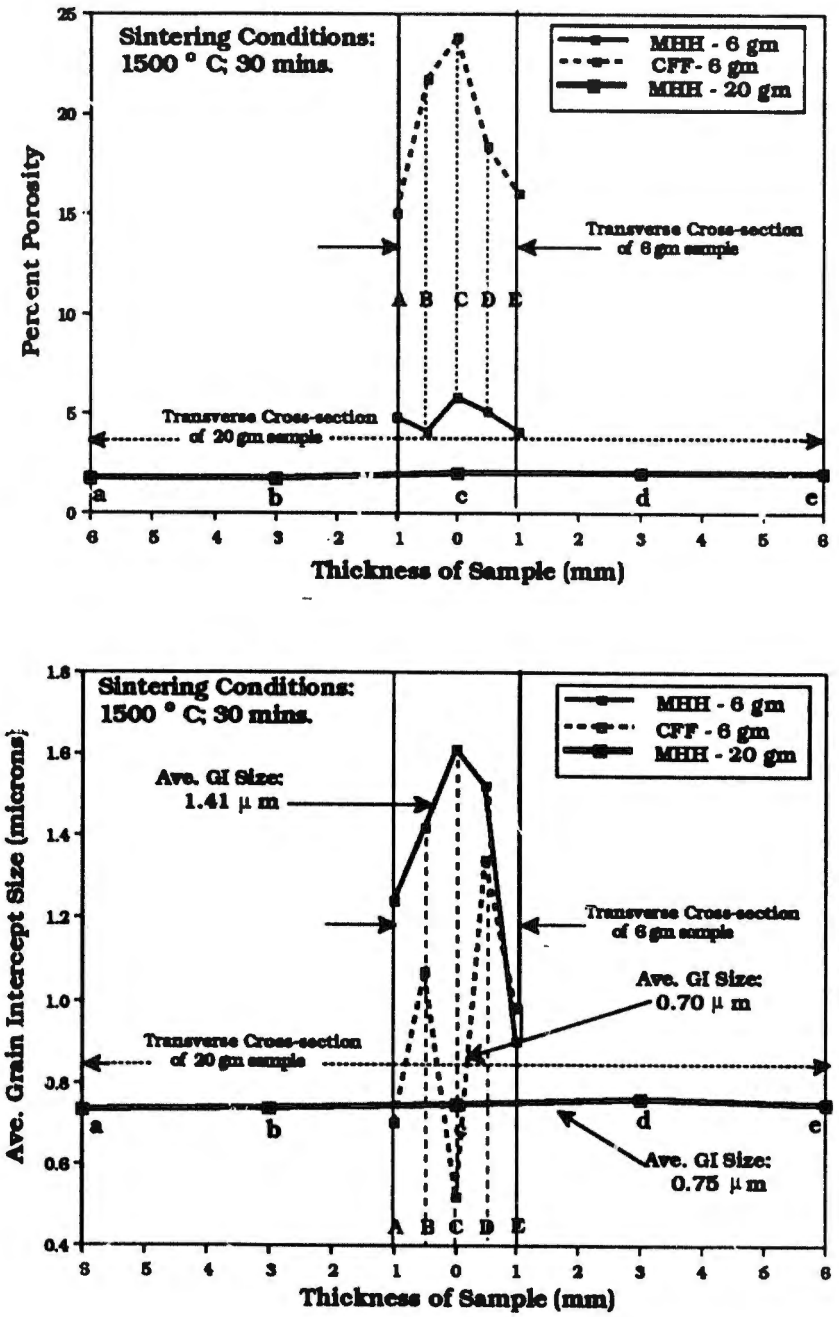

Figure 5. 6 gm (MHH and CFF) and 20 gm (MHH) samples of M-16 alumina sintered at 1500°C 30 min.
(a) Volume fraction of porosity vs. sample thickness
(b) Grain intercept sizes vs. sample thickness

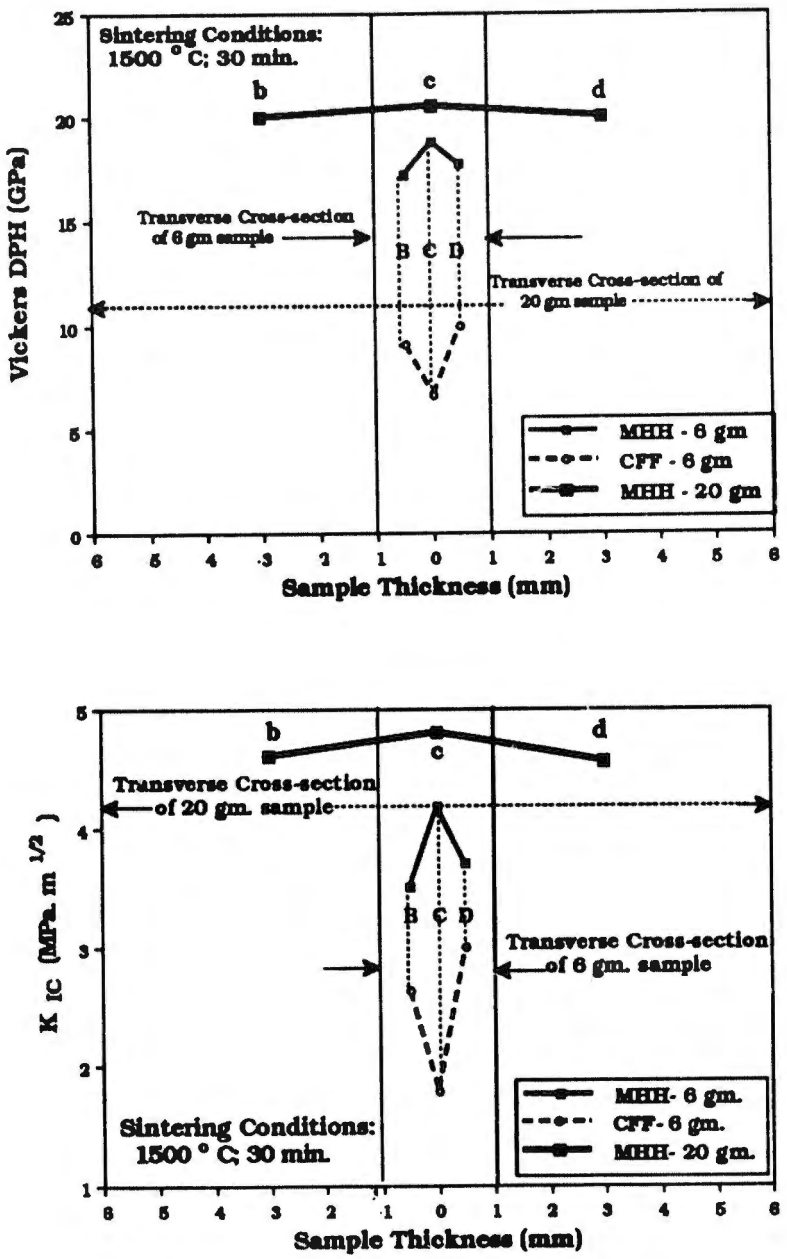

Figure 5. 6 gm (MHH and CFF) and 20 gm (MHH) samples of A-16 alumina sintered at 1500°C 30 min.
(c) Vickers DPN vs. sample thickness
(d) Fracture toughness vs. sample thickness

thickness of the 3 specimens (Figures 5(c) and 5 (d)). Figure 5(c) is a plot of the Vickers microhardness versus sample thickness. The enhanced densification, and uniform grain and pore sizes across the cross-section of the bulk MHH sample is reflected in the enhanced and uniform variation in Vickers hardness numbers (20.06 - 20.54 GPa). The hardness values for the 6 gm MHH sample ranged from 17.26-18.83 GPa, while the variation for the 6 gm CFF sample was of the order of 6.60-9.96 GPa.

Fracture toughness values were extrapolated from diamond indentations, measuring the propagated crack lengths, and computed, using the formulae suggested by Evans and Charles [12]. Trends very similar to those shown by the microhardness numbers are reflected in the fracture toughness (K_{IC}) values. The 6 gm MHH sample had appreciably higher values of fracture toughness (3.5- 4.17 MPa $m^{1/2}$) relative to the CFF sample, with the highest value at the center. The variation for the 6 gm CFF specimen was in the range of 1.8-3.0 MPa $m^{1/2}$, with the lowest values at the interior of the specimen. The 20 gm MHH sample displayed the highest and the most uniform values of the fracture toughness numbers. They ranged from 4.56 to 4.80 MPa $m^{1/2}$, with the highest values at the center of the specimen.

CONCLUSIONS

MW (hybrid) heating facilitates the attainment of perhaps the highest possible heating rates (750°C per minute) attainable with pure, undoped alumina in a 2.45 GHz, untuned, multimode MW cavity.

MHH results in more uniform microstructures and superior properties vs. CFF under the same state conditions of temperature and time. MHH of larger masses result in better volumetric heating, and consequently, an enhanced parity in temperature across the cross-section of the sample. This translates into higher densities, smaller grain sizes, better microstructural homogeneity, and improved mechanical properties. Larger masses, when subjected to CFF, fracture due to large temperature gradients and high thermal stresses across the cross-section.

Enhanced and homogeneous sintering of larger monoliths with MW (hybrid) heating in a 2.45 GHz, multimode cavity is the most significant accomplishment of this research. 'Microstructural uniformity with MW energy', appears to be vindicated with this work. This novel technique for the ultra-rapid sintering of ceramic materials using MW energy holds tremendous promise for the fabrication of advanced ceramics, composites, and bulk monoliths with uniform microstructures, and consequently, enhanced mechanical properties and reliability versus SMW sintering and CFF.

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