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Microwave Sintering of Ceramics for Dentistry: Part 1

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Abstract

The objective of this study was to examine the feasibility of sintering dental ceramic (e.g., zirconia) in a microwave oven, and compare the mechanical properties of these with similar materials sintered in a conventional furnace. Zirconia cylinders were heated to 1100°C, 1300°C, 1350°C, 1400 and 1450°C in microwave and conventional furnaces and sintered at varying dwell times. Our results showed that the mechanical and microstructural properties of samples sintered by microwave were comparable to those of the conventionally sintered samples. Indentation hardness and fracture toughness were found to be 1256 ± 7 and 6.4 ± 0.4 Mpa(m)$^{0.5}$ respectively. The microwave samples were sintered in significantly less time had less voids and more uniform grain structure. Our results suggest that microwave sintering can produce rapid and reliable processing of complex dental ceramics with better microstructural properties and energy savings.

Keywords: Ceramics; Porcelain; Microwave; Sintering

Introduction

During microwave sintering, microwaves penetrate the material; some of the energy is absorbed by the material and converted to heat. The formation of heat raises the temperature of the material. However the surface of the material loses more heat to the surroundings than the interior, hence the interior parts of the material become hotter than the exterior [1].

Microwave Hybrid Heating (MHH) is a process of heating electrically transparent (low-loss) materials like silica, alumina and zirconia [2]. To enable microwave penetration and heating, these samples have to be preheated by another source, which can couple with microwaves at room temperature. The ceramic, silicon carbide is usually used as a susceptor or a pre-heater because it couples well with microwaves at room temperatures unlike other ceramics, e.g. zirconia. Heating is initiated by the coupling of susceptors with the microwave field at room temperature and as the temperature increases the transparent material becomes more susceptible to microwave energy. At a critical temperature, the low loss material becomes more microwave absorbent thus can be heated by microwave.

In recent years, zirconia has become a material of choice for dental restorations [3]. The objective of this study was to demonstrate the feasibility sintering zirconia-using MHH and to compare the resultant microstructural and mechanical properties with those of the conventionally sintered samples as previous studies with respect to the use of microwaves for dental ceramics are lacking [4].

Materials and Methods

Powders of zirconia stabilized with 3 mol % yttria (TOSOH, Japan) were uniaxially pressed (compacted) into cylinders with a radius of 8.36 ± 0.89 mm and height of 5.33 ± 0.34 mm (wt. 0.58 g) using a standard Carver hydraulic piston press and commercial tool steel dies at 2000psi. The samples were cold isostatically pressed at 15000 psi. Thirty samples were utilized for conventional sintering and thirty four were sintered with microwave. Each sample was weighed, and the lengths and diameters were measured with a micrometer (Mitutoyo, Japan). A Thermolyne 5000 series tube furnace was utilized to conventionally sinter 30 samples. The furnace was heated at approximately 10°C per minute to 1450°C. Six cylinders were sintered at a time. A total of thirty cylinders were sintered to 1450°C in this manner utilizing the same ramp rate (10°C/minute). An additional lot of pellets measuring 2.5 mm × 0.4 mm × 6 g were prepared by the above mentioned methods and sintered in Lindberg conventional furnace at approximately 40°C per minute in order to study the effect of increased heating rate.

A multi-magnetron 2 kW commercial research microwave oven having a stainless steel cavity and equipped with dual mode stirrers, a platinum sheated S-type thermocouple and Omega controller was purchased from the Microwave Research and Applications, Inc (Model number BP 210/211) for the microwave sintering experiments. The operating frequency was 2.45 GHz. A thermal and a fibrous alumina cylinder containment box were made using fibrous alumina boards 1.5 inch thick. Three Silicon carbide rings (25 mm diameter) were used as preheaters or susceptors. These were sandwiched between the reticulated zirconia plates which also acted as the floor upon which the samples rested. Peak sintering temperatures of 1100°C, 1300°C, 1350°C, 1400°C, 1450°C, respectively, were used. Two stainless steel pedestals with equally spaced 4 mm holes drilled perpendicular to the vertical axis were utilized to hold the S-type thermocouple. An 8 mm hole was drilled horizontally into the side of the cylindrical portion of the thermal containment cylinder to allow measurement of the temperature within the cavity of the thermal containment box. Temperatures were recorded manually every two minutes. The microwave furnace was started at minimal power. The power was then gradually increased as temperatures ramp dictated until sintering temperature was achieved. All sample-sintering cycles were begun at room temperature.

Post-sintering measurments

Only those sintered cylinders that had a dimensional density of greater than 80% of theoretical (using 6.06 g/cm^3 for zirconia) were analysed using the Archimedes method. The specimens which were sintered at different temperatures were polished and Vickers hardness and toughness values were measured at 20 and 30 kgs.

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The samples from both conventional and microwave sintering were thermally etched to 100°C less than the sintering temperature and held for 1 to 1.5 hours. The samples were sputter coated with gold palladium for 3 minutes (300 angstrom thick coating) and examined by a Scanning Electron Microscope (PHILIPS).

Results

For conventional sintering, 10°C per minute heating rate resulted in a uniform temperature-time profile curve. The sintering temperature of 1450°C was reached in approximately two and half hours. The cooling ramp rate was also nearly 10°C per minute. This heating profile resulted in dense sintered sample with a translucent sheen indicating that they were fully sintered after a period of six hours. Faster ramp rates (in excess of 40°C) rendered insufficient sintering of samples and these specimens were highly porous.

To reach a peak temperature of 1100°C in a M/W furnace, we used a ramp rate in excess of 100°C/min. However, upon removal of the samples after cooling, they were not completely sintered, and had warped to a certain extent. The low temperature of 1100°C with no dwell time was insufficient to sinter the samples and attempts to use dwell times of 20 and 30 minutes resulted in damage to the floor of the microwave oven. The maximum power required to achieve the sintering temperature of 1300°C was 78 percent. For samples that were sintered to 1400°C and 1450°C the power had to be manipulated initially until the temperature rose to about 380°C. Final power of 72 percent increased the microwave heating to an extent that heating rates of approximately 150°C allowed the final temperature of 1400°C or 1450°C to be achieved as shown in Figure 1. Again, the samples, upon examination were intact with no signs of cracking and had a translucent sheen, which was not observed, at other sintering temperatures. Ramp rates in excess of 150°C resulted in damage to the floor of the microwave oven. However, the samples were completely sintered in less than 10 minutes.

At lower microwave sintering temperatures of 1100°C, 1300°C and 1350°C the density was considerably less than the conventionally sintered samples. There was no significant difference in the density at 1450°C for both microwave and conventionally sintered samples as shown in Table 1. The comparison of hardness between the conventionally sintered and the microwave sintered samples at 30 Kg load yielded similar results with the samples sintered with microwaves at 1450°C displaying higher hardness values than the conventionally sintered samples at the same temperature. The fracture toughness increased with the sintering temperature as shown in Table 1 and the values for HV_{20} indentations were comparable with the microwave-sintered samples at sintering temperatures 1450°C as with those samples sintered conventionally at that temperature. Cracks measured at the center had propagated more than those measured at the edges of the samples. The average microstructure of the microwave-sintered samples had less voids than that of the conventionally sintered samples when examined at the same magnification of 10000X. No significant difference in the grain shape was observed between the two sintering methods. The samples sintered in a conventional furnace with slow ramp rate of 10°C per minute had slightly larger grains in the interior than the exterior as shown in Figure 2. The average grain size at the exterior was approximately 0.3 μm, while at the interior; it was approximately 0.7 μm. This suggests that the temperature of the external surface might have been higher than the interior. Few voids were also seen in these samples. The microstructure as shown in Figure 3 appeared to be more uniform throughout the sample and few slightly larger grains (0.5 μm)

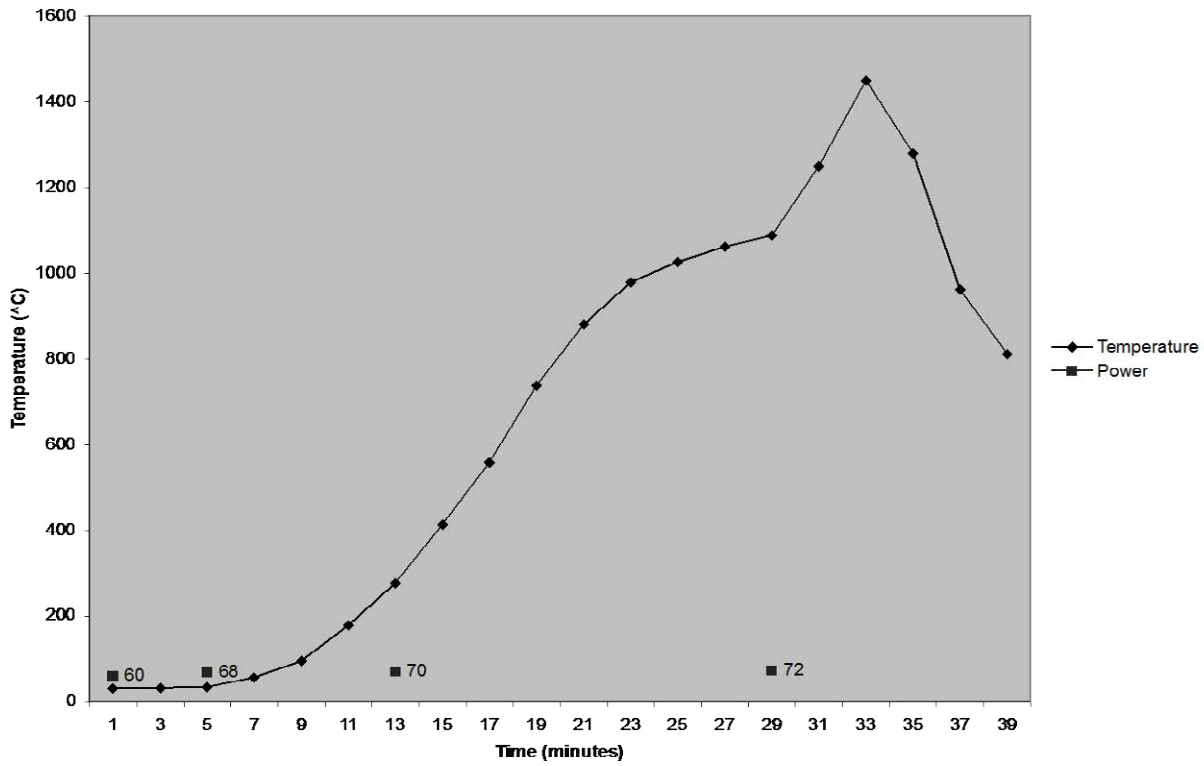
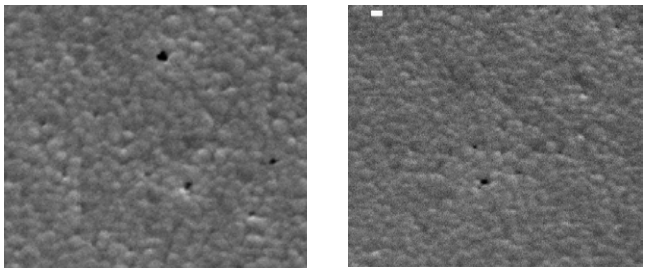


Figure 1: Time vs temperature profile along with microstructres of a zirconia sample sintered in a microwave. The percentage of full power utilized is also shown along with the changes in microstructure.

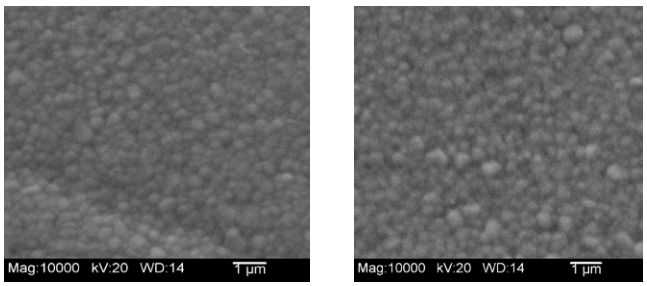
Temp (°C)	VHN_{20}	Kic_{20} (Mpa. $m^{0.5}$)	VHN_{30}	Kic_{30} (Mpa.$m^{0.5}$)	Density
1100-m/w	206 0.7	3.1 ± 0.2	275 ± 7.8	Inaccurate	56.3 ± 3.7
1300-m/w	827 ± 16	4.8 ± 0.13	863 ± 45	5 ± 0.23	90 ± 2
1350-m/w	994 17	5 ± 0.4	895 ± 7	5.5 ± 0.3	93 ± 0.2
1400-m/w	1039 ± 11	5.6 ± 0.2	1035 ± 10	6.4 ± 0.2	96 ± 0.9
1450-m/w	1256 ± 7	6.4 ± 0.4	1267 ± 18	6.8 ± 0.13	98 ± 0.9
1450-Conventional	1130 ± 25	6.5 ± 1.1	1237 ± 29	6.6 ± 0.8	98.04 ± 2.37

Table 1: Mean standard deviation of the Mechanical Property (Vickerns Hardness Numbers (VHN at 20 and 30 kgs) and respective fracture toughness (Kic at 20 and 30 kgs)) of Zirconia samples sintered in a microwave at various temperatures and in a conventional oven at 1450°C.



A B

Figure 2: SEM micrographs displaying smaller grains in the exterior (B) as compared to the interior (A) of the conventionally sintered samples (1450°C, 10000X).



A B

Figure 3: SEM micrographs showing uniform grain size distribution for microwave sintered samplesin both exterior (A) and also in the interior (B) due to volumetric heating (1450°C, 10000X).

interspersed with the densely packed smaller grains (~ 0.3 µm) with the average grain size measured to be around 0.3 µm.

Discussions

High strength ceramic materials that are inexpensive and durable when subjected to cyclic loading in oral environment and can be made into individually constructed restorations have become available in the market [5]. Such zirconia or alumina blanks for fabricating dental crowns are normally sintered or pre-sintered in a conventional oven. We mimicked the heating and cooling rate employed by a normal dental laboratory for sintering the zirconia blanks in a conventional oven. Conventional sintering with faster ramp rates of about 35°C per minute was attempted. However, faster ramp rates rendered insufficient sintering of samples and consequently were highly porous. A possible explanation for the warping may be attributed to uneven temperature distribution for conventional sintering. In contrast, when faster heating rates (40°C) were applied to the conventional sintering, the samples warped or were distorted due to incomplete sintering. This may be the main reason as to why the dental furnaces for sintering of the ceramic core adhere to slower heating rates. The thermocouple measured the temperature contained within the thermal containment box close to the actual sample and not the actual surface temperature of the samples itself [6-10]. Also, it should be noted that microwave heating is a volumetric heating phenomenon resulting in the interior of the samples being at a higher temperature then the surface [2].

The silicon carbide susceptors (rings) due to their high susceptibility to microwaves at room temperature coupled well with the microwaves and resulted in transformation of this energy to infrared energy which was emitted as heat. Heat began to build up in the containment chamber, which in turn increased the temperature of the ceramic samples. The rate of heating was sufficiently high to achieve high temperatures, which were effective in the coupling of the microwave energy with the zirconia (both the reticulated zirconia and the sample) at the critical couple-triggering temperature. Zirconia, at this point, in a given sintering process, became more susceptible to microwave energy, thereby absorbing energy at a rate greater than that of the silicon carbide susceptors.

This study showed that the most significant factors affecting the density of samples in microwave and conventional heating were the sintering temperatures. Increasing the ramp rate made no significant difference in the density-temperature relationships between conventionally and microwave sintered samples. The conventionally sintered samples with heating rates of 10°C per minute compared well with the faster (>90°C) heating rates of microwave-sintered samples. The hardness values of the microwave samples sintered at 1450°C were comparable with that of the published values for conventionally sintered zirconia when used as a dental biomaterial [5].

HV_{20} indentations were comparable with the microwave-sintered samples at sintering temperatures 1450°C as with those samples sintered conventionally at the same temperature. The increase in the standard deviation of the hardness values for conventionally sintered samples may be due to the non-uniformity in the grain size. Because the surface energy for diffusion mechanism is from the exterior to the interior, the grains at the edges were considerably smaller than those at the centers as shown in Figure 2, which absorbed more energy absorption when indented and may have prevented further propagation of the crack. Moreover, the amount of transformed grains, or the size of transformation zone depends upon the transformability of the tetragonal phase, which is controlled mainly by the grain size [11]. Also, the fracture toughness values are influenced by the mode of propagation of the cracks [3]. The indentation fracture toughness values for samples sintered in a microwave oven or in a conventional oven at 1400°C were comparable (Table 1).

Mechanical properties of a material are not only influenced by its microstructure, but also by the defects present [12,13]. The uniform microstructure as shown in Figure 3 was more uniform throughout the sample perhaps due to the volumetric heating by microwave.

Conclusion

Summarily, transfer of new processing technology from the ceramic industry to dental materials has realized the potential of microwave sintering for dental ceramics. The study suggests that further investigations of the role of the dwell periods at sintering temperatures of 1250-1300°C should be conducted. Sintering at lower temperatures would provide more energy savings and may need less furnace time. The purpose of this study was to examine the feasibility of microwave

ovens in sintering dental ceramics and to develop an ideal microwave sintering profile for firing of crowns and bridges, which could be used in restorative dentistry. The results of this study demonstrate the enormous time savings and other benefits of very rapid sintering. We hope that based on our work new industries related to microwave sintering of dental ceramics would be developed.

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