Variations in Hardness and Grain Size with Density for Fully Stabilized Zirconia Sintered Using Microwave Processing

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Abstract

Densification of ceramic materials with microwave energy is being considered as an alternative means to conventional methods. Using microwave processing for sintering ceramics, many researchers have observed higher densities at lower temperatures, as compared with conventional sintering methods. Microwave sintering technology is under investigation for fabricating inert matrix materials that would recycle fuel in proposed Generation IV nuclear reactors. This study examined the variation in hardness with density and the changes in microstructure with processing technique and temperature. The 8 mol% Y_2O_3-ZrO_2 samples selected for this study were sintered using a conventional furnace, a multi-mode microwave furnace and a single-mode microwave furnace. Microstructural results on 96% theoretically dense samples showed differences in grain size to processing technique (conventional = 1.58 μm, multi-mode microwave = 1.29 μm, and single-mode microwave = 8.86 μm). Vickers hardness tests were performed on select sample groups representative of each processing method. Similarly, dense samples sintered with a conventional furnace yielded a hardness of 5.37 GPa. Whereas, higher hardness values were observed in similarly dense samples that were processed in a multi-mode microwave (9.28 GPa) and single-mode microwave (7.77 GPa). The results obtained in this study favor microwave sintering at low temperature that could eventually find an application in fabricating inert matrix fuels for next generation reactors.

Keywords: Zirconia, Microwave Processing, Hardness

1. Introduction

Inert matrix fuels are being developed to recycle the spent nuclear fuel in the proposed Generation IV nuclear reactors[1-3]. These newly developed reactors will use the spent nuclear fuel, which is a byproduct of existing reactors. The problem with the radioactive materials in spent nuclear fuel (more formally known as transuranic nuclides) is that they are volatile at high temperatures (1600 °C). This behavior will result in material loss when using high temperature conventional processing. Microwave processing is being explored to fabricate these fuels at lower temperatures[4].

Extensive research has been done to determine the materials that are best suited for use as an inert matrix[5]. The materials must achieve a minimum strength (20-30 GPa) to withstand the high pressures experienced by a fuel pellet during its normal operation. They must also maintain a minimum percentage of residual porosity to contain the daughter products of the fission reaction. The ideal densification to satisfy these parameters is 85-95% theoretical density. Apart from hardness, structure (grain size) plays an important role in releasing the fission gasses that are a byproduct of a nuclear reaction[6]. Stabilized zirconia is a candidate material that has been found to have the necessary traits for this application[5].

The objective of this work is to study the hardness achieved in samples that have been processed in a conventional furnace and the two different microwave furnaces. Also, another objective is to observe the microstructural differences seen as a result of the different processing techniques.

2. Experimental Procedure

2.1 Materials and Preparation

Pellets of 8 mol % Yttria-Zirconia (8YZ) were formed using a combination of uniaxial and isostatic pressing. Sintering was performed in a conventional furnace and two different microwave furnaces (multi-mode microwave (MMM) and a single-mode microwave (SMM)). Microwave sintering results showed higher percent theoretical densities at lower temperatures[7]. A theoretical density value of 8YZ powders used in this study was estimated by R. R. Thridandapani et al. to be 5.96 g/cc (this value was obtained from the X-ray diffraction patterns).

Samples were sectioned from sintered pellets of 8YZ. The population consisted of samples processed at different temperatures (1100, 1200, 1300, 1400, and 1500 °C).

A ¼ inch wide sample was cut from the center of each pellet. This section of the pellet was chosen because it allowed more area for hardness testing to be conducted on each sample. They were then mounted, ground and polished to prepare them for hardness testing. Silicon carbide pads with grit sizes of 180, 240, 320, 400, 600 and 1000 were used for grinding. Polishing was done on felt pads using 15, 6, 3, and 1 micron diamond polishing suspension.

2.2 Vickers Hardness

Indentations were created using a Vickers micro indenter at a force of 9.8 N for a dwell time of 5 seconds. Figure 1 shows a typical view of a Vickers indentation.

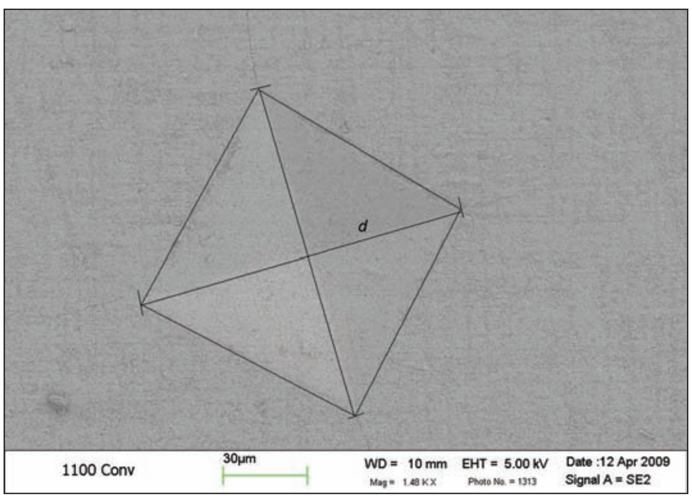

Figure 1. An example of a Vickers indentation (outlined in black).

The tests were conducted according to ASTM C 1327 standards and hardness values were calculated using equation 1.

$$HV = 0.0018544\ P/d^2 \qquad (1)$$

Where HV is the Vickers hardness value, P is the indenter load in newtons, and d is the length of the diagonal in millimeters[6].

2.3 Determination of Grain Size

For this study, the most easily measured microstructural feature is grain size. After gathering hardness data, each sample was prepared for Scanning Electron Microscopy (SEM) through thermal etching. During hardness testing, the surface of the sample must be highly polished. Therefore, due to the necessary surface etching, the SEM micrographs must be taken after the hardness testing is completed. SEM micrographs were taken for several samples from each temperature and sintering method.

Grain size measurements were conducted using the intercept method according to ASTM standards E 112. The average grain diameter was calculated using equation 2.

$$\bar{l} = L_T/N_i \qquad (2)$$

Where $\bar{l}$ is the "mean lineal intercept," which can be interpreted as the average grain diameter of the sample in µm, L_T is the test line length in µm, and N_i is the number of intercepts per test line[9].

3. Results and Discussion

3.1 Variations in Hardness with Density

As the density of the samples of 8YZ increased to approximately 96% theoretically dense, the Vickers hardness increased as well. For 96% theoretically dense specimen, the processing technique used (Conventional Furnace, MMM, or SMM) did not show a significant difference between hardness values. Figure 2 illustrates these points for the 8YZ samples processed using a conventional furnace, a multi-mode microwave furnace and a single-mode microwave furnace.

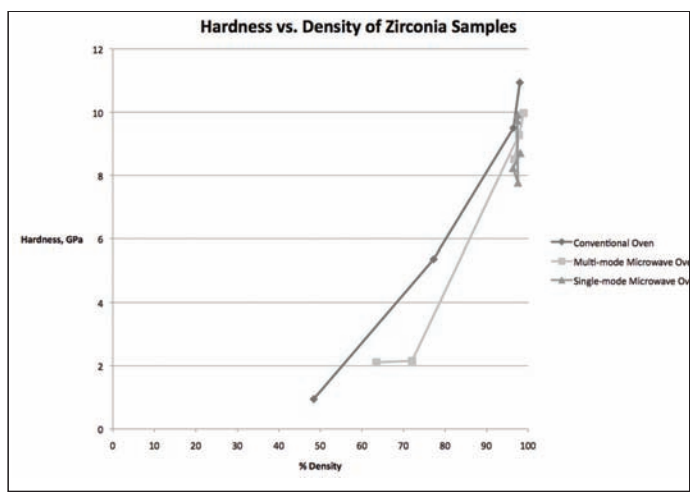

Figure 2. A plot of hardness vs. percent theoretical density for 8YZ.

3.2 Variations in Grain Size with Sintering Temperature

The samples sintered at 1100° and 1200° C using a conventional furnace and a MMM furnace did not reach full density. The microstructure of these samples showed no significant differences between each other.

All other samples were at least 96% theoretically dense, except the sample sintered at 1300 °C processed using the conventional method. This sample had a density of about 77% theoretical. The variation in sintering temperature with grain size is shown in Figure 3. It can be observed that grain size increased as the sintering temperature increased.

The data represented in Figure 3 also shows that the samples sintered using a single-mode microwave experienced a significant increase in grain size with temperature. It has been reported by many researchers that the diffusion is enhanced due to the presence of microwave energy[10]. The observed increase in grain size may be due to this improved diffusion occurring in the SMM sintering process.

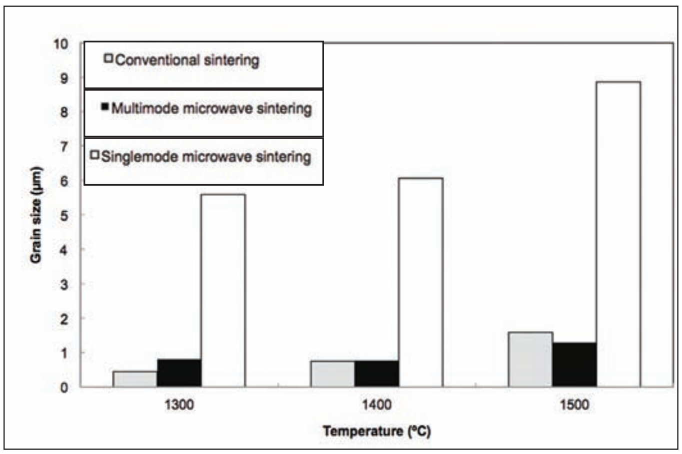

Figure 3. A bar chart of the grain size at different processing temperatures for 8YZ samples.

The micrograph images in Figure 4 show the grains and their boundaries that exist for fully dense samples of 8YZ sintered using the three different methods. An important point to notice is the similarity between the conventionally sintered and the MMM sintered samples. It can also be observed that SMM sintered samples showed a larger grain size than those produced using the other two processing techniques.

4. Conclusions

In conclusion, the hardness values showed a significant increase with percent theoretical density. The results also showed that higher hardness was achieved at lower temperatures in a microwave furnace, indicating that this method could be useful for fabricating inert matrix fuels at lower temperatures with less material loss without compromising hardness.

It was observed that the SMM samples showed a significant increase in grain size with temperature when compared with samples processed using a conventional furnace and a MMM at the same temperature. The result implies that nuclear fuels which demand larger grain size could be processed at much lower temperatures in a SMM furnace.

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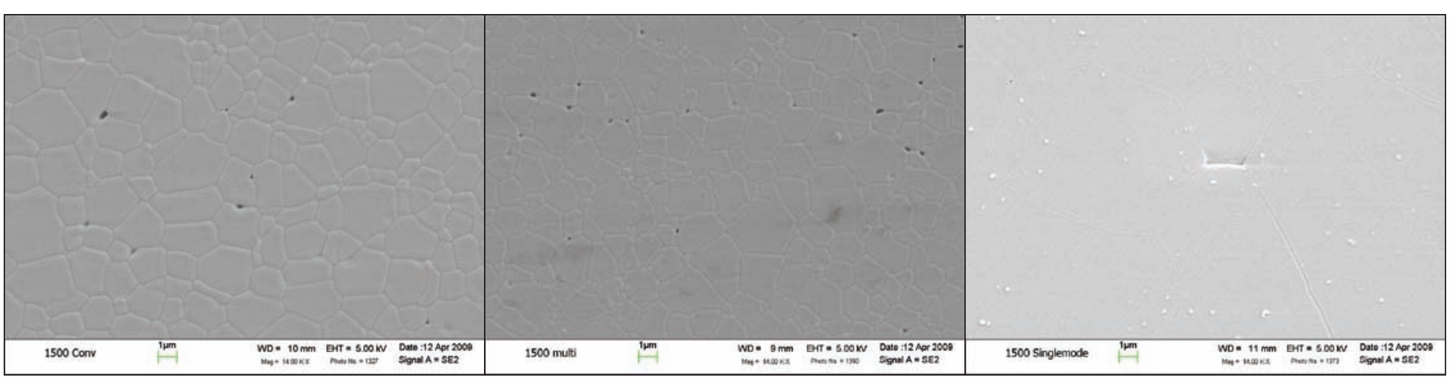

Figure 4. Micrographs showing the grains for conventionally sintered (left), MMM (center), and SMM sintered (right) samples of 8YZ; T = 1500°C.

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About the Author

Andrew is a senior in the Department of Materials Science and Engineering at Virginia Tech. He is on track to receive an official certification in nuclear engineering from the Department of Mechanical Engineering with his Bachelor of Science degree. He was a research assistant in the Microwave Processing Research Facility, under Dr. David Clark and Diane Folz, during the Spring and Summer of 2009. He is primarily interested in the properties, processing and structure of ceramic materials, and is planning to pursue a Master of Science degree in MSE which will focus on research in ceramic materials.