COMPARISON OF MICROWAVE AND CONVENTIONAL SINTERING OF Al_2O_3-ZrO_2 COMPOSITES

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1. Introduction

Oxide ceramic materials was used in variety of modern technological process due to a unique combination of physicochemical properties. Alumina has an excellent property in high temperature stability, high strength, high hardness, biological resistance, thermal stability, and chemical resistance. It is widely used for various applications such as spark plug, ball mill and pot mill, electronic substrate, and etc [1]. Zirconia exists as a monoclinic crystal at room temperature and inverts to tetragonal phase above approximately 1200°C. It has high density, high fracture toughness, high hardness, low thermal conductivity, chemical inertness, and ware resistance [2]. Al_2O_3-ZrO_2 composite consists of an alumina matrix in which there are embedded zirconia particles, either unstabilized or stabilized. It is well known that this second phase addition results in an enhancement of their mechanical properties such as high strength, and high toughness. Due to their excellent properties, Al_2O_3-ZrO_2 composite remains an interesting subject for materials researchers and has been used in various application in recent year [3]. Microwave sintering has gained increasing attention to scientist because of its advantages over conventional sintering for ceramic materials. Microwave belong to electromagnetic spectra with wavelengths from 1 mm to 1 m. The commonly used frequency for microwave heating are 2.45 and 0.915 GHz [4]. In conventional heating, energy transferred to the materials through convection, conduction, and radiation of heat from surfaces of the material. In microwave heating, energy is delivered directly to materials through molecular interaction with the electromagnetic field. This difference of energy delivery can result in many advantages of microwave heating such as uniform heat, rapid heating rate, short cycle time, higher toughness and fine grain size, higher mechanical properties, higher density, lower firing temperature. However, very little literature reported on the microwave heating of Al_2O_3-ZrO_2 composite. The aim of this paper is to present a comparative study of the microwave heating and conventional heating applied to Al_2O_3-ZrO_2 composite in related to their properties of density, porosity, and strength.

2. Experimental Procedure

Starting materials used to prepared the samples are high purity Al_2O_3 (98.9%) with average particle sizes of 3.2 μm and fine $ZrSiO_4$ powder with average particle sizes of 0.3 μm and 9.7 μm. Two batch compositions were prepared, AZ1: 80%wt Al_2O_3 + 20%wt $ZrSiO_4$ (0.3 μm) and AZ2: 80%wt Al_2O_3 + 20%wt $ZrSiO_4$ (9.7 μm). The mixture of each batche was wet milled for 5 h, dried and sieved. The green pellets were formed by uniaxial pressing at 2 tons. The dimensions of green pellets were 1.23 cm in diameter and 0.35 cm in thickness and dimension for the bar shape was 1 x 5.5 x 0.7 cm. The firing profile was 5°C/min up to 600°C and then 10°C/min up to 1300, 1400 and 1500°C, respectively. The bulk density, porosity and water absorption were measured by the Archimedes' method. The flexural strength were determined from three-point bending test with a span length of 30 mm and loading rate of 0.5 mm/min. Phases of samples after firing were characterized by X-ray diffraction, with Ni filtered Cu$K\alpha$ radiation (XRD:Shimadza, Japan). Microstructures were observed from Scanning electron microscopy (SEM: JOEL JSM-6340F).

3. Results and Discussion

Typical X-ray diffraction pattern of samples (AZ1) after sintering at 1300°C and 1400°C in both conventional and microwave have been presented in Fig. 1 and 2, respectively. It was found that sintering at 1300°C in conventional furnace, α-Al_2O_3 and $ZrSiO_4$ were observed as major phases while the monoclinic zirconia (m-ZrO_2) showed as minor

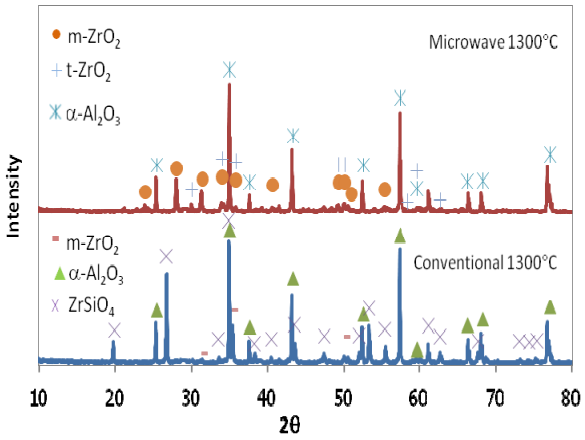

Fig. 1. XRD pattern of AZ1 sintered at 1300°C

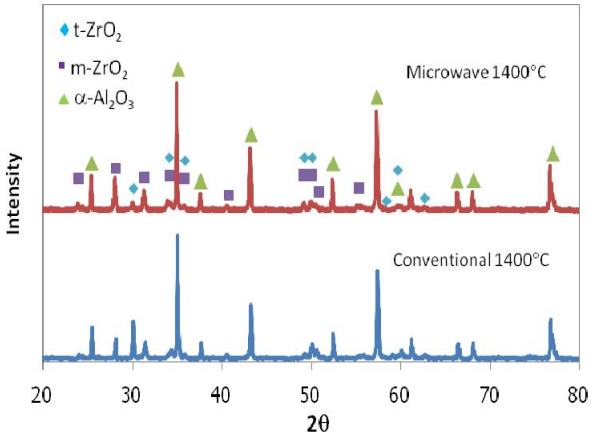

Fig. 2. XRD pattern of AZ1 sintered at 1400°C

phase and the tetragonal zirconia (t- ZrO_2) was not observed (Fig.1). By increasing sintering temperature to 1400°C, higher intensity peaks of m-ZrO_2 and t- ZrO_2 were observed and peaks of $ZrSiO_4$ were rarely appeared. For samples sintered in microwave furnace, both m-ZrO_2 and t-ZrO_2 were found at temperature 1300°C and 1400°C and intensity of peaks increase with increase temperature. However, $ZrSiO_4$ peaks were not observed for both sintering temperature in microwave furnace. This can be explained that during sintering $ZrSiO_4$ will transform to m-ZrO_2 and to t-ZrO_2 at higher temperature. By microwave sintering, the volumetric interaction of the electromagnetic fields with a ceramic material will lead to a higher heating efficiency and faster reaction rates when compared with conventional heating at the same temperature [5, 6]. This resulted in more ZrO_2 phase formed in the sample sintered by microwave.

The results of bulk density, apparent porosity and water absorption of AZ1 and AZ2 samples sintered at 1300, 1400 and 1500°C in conventional and microwave furnaces were shown in Fig. 3, 4 and 5, respectively. AZ2 sample sintered in conventional furnace at 1300°C presented the lowest density of 2.4 g/cm^3, highest porosity and water absorption of 42 and 17.5%, respectively. This high porosity is caused from densification retarding of bigger particle size of $ZrSiO_4$. However, this phenomenon can be compensated by using microwave sintering. Microwave sintering process can produce higher final density of sample than the conventional sintering process. It can enhance densification, especially at higher sintering temperature presents more evident effect. This is because microwaves absorb the electromagnetic energy volumetrically, and transform it into heat which generate within the materials first and then transfer to the entire volume. This is different from conventional heating, in which heat is transferred between particles by the mechanisms of conduction, convection and radiation, the material's surface is first heated followed by the heat moving inward. This means that there is a temperature gradient from the surface to the inside.

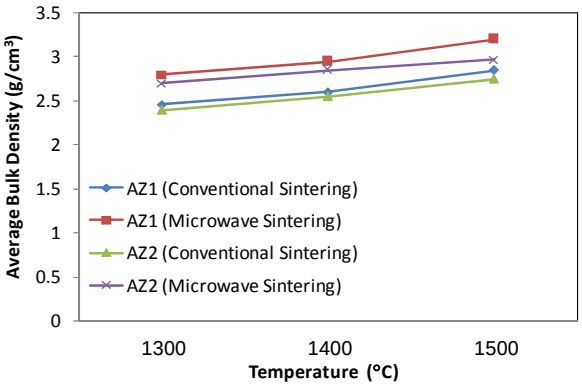

Fig. 3. Average bulk density of AZ1 and AZ2

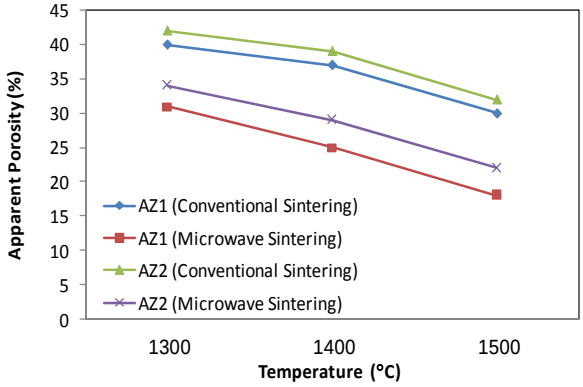

Fig. 4. Apparent porosity of AZ1 and AZ2

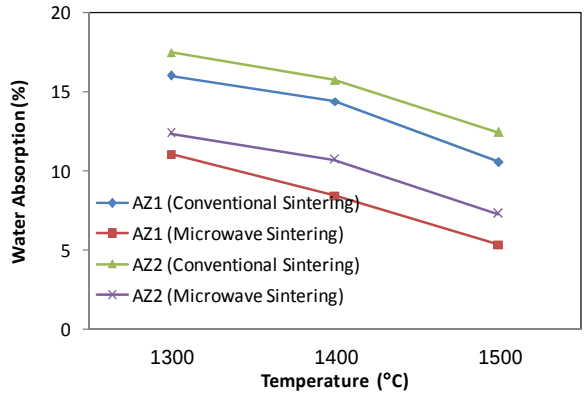

Fig. 5. Water absorption of AZ1 and AZ2

The results of flexural strength and compressive strength from the effect of $ZrSiO_4$ starting particle sizes, sintering temperature and furnaces were shown in Fig. 6 and Table 1. It was found that flexural strength and compressive strength of sintered samples increased significantly with increasing sintering temperature under the same particle size and furnace. AZ1 sample performed higher flexural and compressive strength compared to AZ2 sample sintered at the same temperature and furnace. It was found that flexural and compressive strength increase with decreasing starting particle sizes. Samples prepared from smaller particle size performed higher flexural and compressive strength than those prepared from larger particle size under the same sintering temperature and furnace. This is due to small particle size creates high density of sintered body which results in high strength of particle bonding. The composite samples sintered by the microwave method exhibited higher flexural and compressive strength than that samples sintered by conventional one. This improve of strength can be explained by homogeneous microstructure of sample sintered in microwave furnace. This result revealed superior mechanical properties of microwave sintered sample compared to conventional sintered samples. Furthermore, in Al_2O_3-ZrO_2 composite, t-ZrO_2 can be formed at all temperature by microwave sintering.

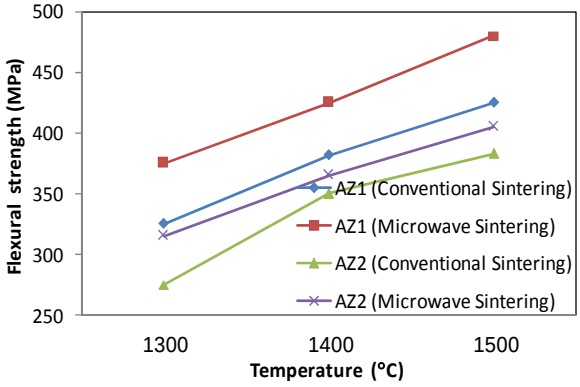

Fig. 6. Flexural strength of AZ1 and AZ2

Table 1. Compressive Strength of AZ1 and AZ2

Sintering Temperature (°C)	Compressive Strength (MPa)			
	Conventional		Microwave	
	AZ1	AZ2	AZ1	AZ2
1300	42	31	48	39
1400	44	35	50	41

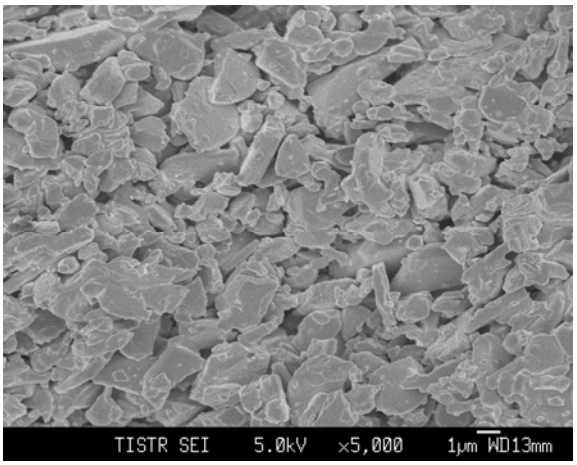

Fig.7. SEM micrograph of AZ1 sintered at 1300°C in conventional furnace.

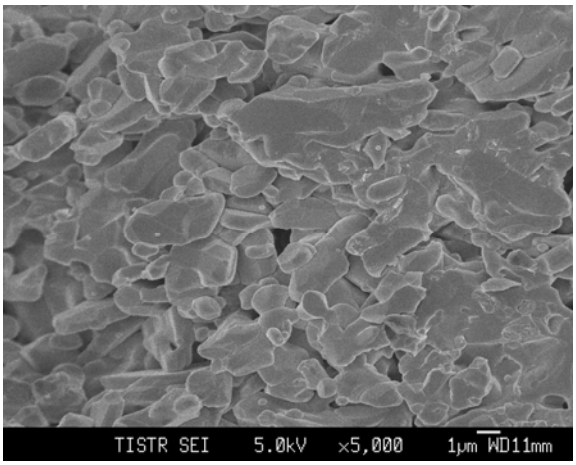

Fig. 8. SEM micrograph of AZ1 sinterd at 1400°C in microwave furnace.

Fig. 7 and Fig. 8 Shows micrographs of fracture surfaces of the two composites, one was sintered at 1400°C in the conventional furnace and the other was sintered at 1400°C in microwave furnace. In case of the conventional sintering (Fig. 7), the grain size appears to be nearly identical to that of the initial particles. The microstructure of the AZ1 sintered by microwave furnace (Fig. 8) presented the enhanced neck growth between initially touching particles and dense microstructure. More grain growth was observed in the sample sintered by microwave at the same temperature (Fig. 9). Microstructure of microwave sintered samples showed large pores but small in numbers whereas smaller pores and more in number for conventional sintered samples (Fig. 7 and 8). The microstructure shows the decreasing of porosity with increasing sintering temperature (Fig. 8 and 9). Microwave synthesis of materials is fundamentally different from the conventional process in terms of its heating mechanism. In a microwave furnace, heat is generated within the sample volume itself and energy heats the materials on a molecule level, which leads to uniform heating, which results in homogeneous and dense microstructure.

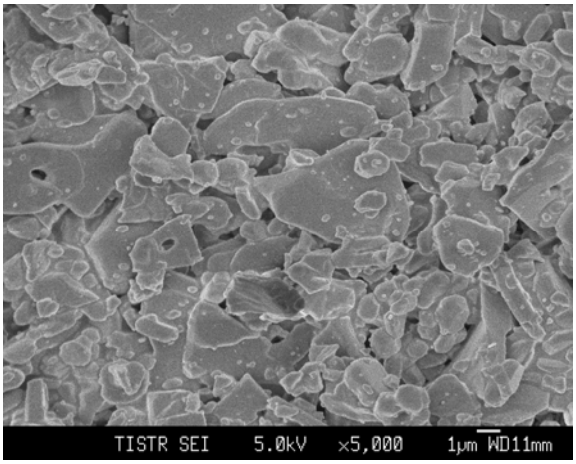

Fig. 9. SEM micrograph of AZ1 sinterd at 1300°C in microwave furnace.

4.Summary

Microwave processing is generally believed to be method of highly efficient production. The two

different sintering process (microwave and conventional) for Al_2O_3-ZrO_2 composite produce different phase transformation, microwave sintering promoted the formation of t-ZrO_2 at 1300^oC and 1400^oC, while this phase forms in sample with conventional sintering at 1400°C. Microwave sintering results in higher densities, flexural and compressive strength compared to conventional sintering. The porosity of sintered Al_2O_3- ZrO_2 composite increased with increasing of starting $ZrSiO_4$ particle size. Furthermore by microwave sintering, the microstructure of composites show more dense and uniform grain growth.

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