Volumetric Microwave Heating of Mullite Ceramic Using a 28 GHz Gyrotron

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Abstract: In this paper, we present and discuss experimental results from a microwave sintering of a mullite ceramic, produced from a mixture of alumina and silica xerogel extracted from a sago waste ash. The composition has been prepared by adding 60 %wt amount of alumina into the silica xerogel. As a radiation source for the microwave heating, a 28 GHz wave gyrotron has been used. The influence of the sintering temperature on the structural properties was studied in detail. It has been found that the mixture crystallizes completely as a single mullite phase at a temperature of 1600 °C after heating for 10 min at a temperature rate of 45 oC/min. Most importantly, such result cannot be achieved by conventional heating. The results that have been obtained in this study allow one to conclude that the microwave sintering of using a 28 GHz gyrotron is an appropriate technological process for the production of mullite ceramics and is characterized by several advantages such as shorter times of the thermal cycle, lower sintering temperatures and higher quality of the final product.

Keywords: Alumina, silica xerogel, sago waste, mullite, microwave heating, gyrotron

1. Introduction

Mullite is an excellent material and has numerous uses in various industrial structural and functional applications due to its unique properties. For example, it has extremely high-temperature stability, thermal shock resistance, resistance to chemical attack and abrasion which is useful for its usage as a refractory material [1]. The low coefficient of thermal expansion, low density and low thermal conductivity make mullite an appropriate material for the production of optical infrared windows [2]. Additionally, its low dielectric constant and conductivity are suitable for fabrication of ceramic substrates for microelectronics packaging [3].

It has already been reported that the formation of mullite in the sample depends on the type of the used precursor gel, particle size and the parameters of the thermal treatment process. Preceding studies in which both the electric and the microwave heating sources have been used have demonstrated the possibility of forming a single crystalline phase of mullite. In order to achieve this, the most important

factor is the heating of alumina and silica at sufficiently high temperature. In the case of an electric or conventional heating, the formation of single mullite phase in the mixtures of $Al(OH)_3$ and micro silica occurs at 1600˚C for 3 h at a heating rate of 3˚C/min [4]. The formation of mullite is intense when the heating rate in the mixture of clay and alumina source is raised up to 5˚C/min [5]. Recently, mullite has been prepared using an alumina and silica xerogel from sago waste as starting powder and the complete mullitation has been achieved at a temperature of 1600˚C for 2 h and a heating rate of 3˚C/min [6]. These studies have demonstrated several disadvantages of the used crystallization process, most notably very low heating rates and long soaking times during sintering, which promote grain growth. Such problems can be solved replacing the conventional heating by a microwave heating.

Inherent advantages of the microwave sintering are significantly higher heating rates and a volumetric heat generation. Additionally, such heating by microwaves significantly diminishes the time for grain growth. In a series of preceding experiments, the mullite ceramic has been prepared by a volumetric microwave heating using a microwave oven operating at a frequency of 2.45 GHz and the results have been compared with the conventional treatment. It has been observed that the mullitization process was completed after 20 min in a mixture of clay and alumina and after 25 min in bentonite clays [7]. Brasileiro et al. (2012) [8] compared the conventional and microwave processing of mullite derived from kaolin residue. They have reported a formation of secondary mullite in just 20 min of sintering in a microwave oven (at a frequency of 2.45 GHz and an output power of 1.44 kW) while the conventional sintering at 1400˚C requires a much longer time of 280 min. This result is supported by the study of mullite formation in clay and alumina composites [9] and kaolinite [10], which shows that the microwave treatment reduces the sintering temperature by 120 and 400 degrees, respectively. In their entirety, these results demonstrate the following advantages of the microwave sintering: (i) a significant mullite densification; (ii) a more uniform structure and increased mechanical strength due to the formation of smaller grains; (iii) an improvement of the reaction rate and, eventually, (iv) an improved quality of the mullite ceramic. In this paper, we present the results of our study on the structural properties of a mullite ceramic sintered by a microwave heating using a gyrotron as a radiation source

2. Experimental Procedure

2.1. Silica Xerogel and α-Al_2O_3 Composite Preparation

Amorphous silica xerogel (SX) was extracted from a sago waste (solid residue, which is left behind after the starch has been washed out) obtained from the sago processing plant in Kendari, Indonesia. The extraction procedure is described in detail elsewhere [11], [12]. The composite powder was prepared mixing such SX with monosized crystalline α-Al_2O_3 powder having a particle size of 300 nm. The preparation procedures of the composite powder are described in the literature [6]. The dried powder and 3 wt% polyvinyl alcohol (PVC) were mixed and then pressed at 20 MPa using a hydraulic compaction to form cylindrical samples with a diameter of 10 mm and a thickness of 7 mm. The ceramic green body samples were dried at 300°C with a drying rate of 1.5°C/min and afterwards were stored in a desiccator for a further processing.

2.2. Sintering of the Green Body of Silica Xerogel and Al_2O_3 Composite

The experiments on the green body sintering have been carried out using microwave heating by gyrotron radiation with a frequency of 28 GHz [13] to temperatures in the range from 1300 to 1700°C. A controlled heating rate of 45°C/min has been maintained up to the desired temperature in the applicator and then the temperature has been kept constant for 15 min. The sample holder was made of heat insulating material (Fibermax). The temperature was measured using an R-type thermocouple placed in contact with the

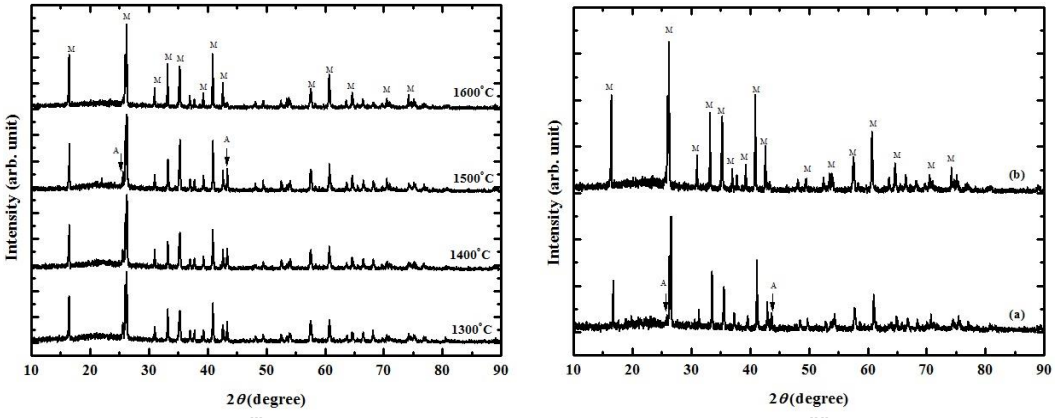
surface of the sample. The cooling was performed by natural convection after turning the furnace off and leaving the samples inside.

2.3. Methods Used for the Structural Characterization of the Mullite Ceramic

The identification of the crystalline phases formed during the sintering was performed by XRD in a Smartlab X-ray diffractometer, supplemented with primary and secondary monochromators and controlled by a computer. The X-ray tube was operated at 40 kV/30 mA with filtered Cu Kα radiation at a wavelength of 0.15418 nm. The instrument was run in a scan mode with an increment of 0.02 and a scan speed of 5 s/step within an angle 2θ ranging from 10 to 90 degree. Furthermore, in this study, micrographs were taken in order to evaluate the characteristics of the surface of the sintered samples. The surface morphology of the sintered samples was then examined in more detail by a scanning electron microscope (SEM) JOEL-6480LV. As the electron beam penetrates the surface of the sample, it produces emission of secondary electrons that are collected by detectors. The signal from the detectors is used to register an image of the surface of the sample on a monitor. The images were registered at a magnification of 5000 times and an accelerating voltage of 15 kV.

3. Experimental Results and Discussion

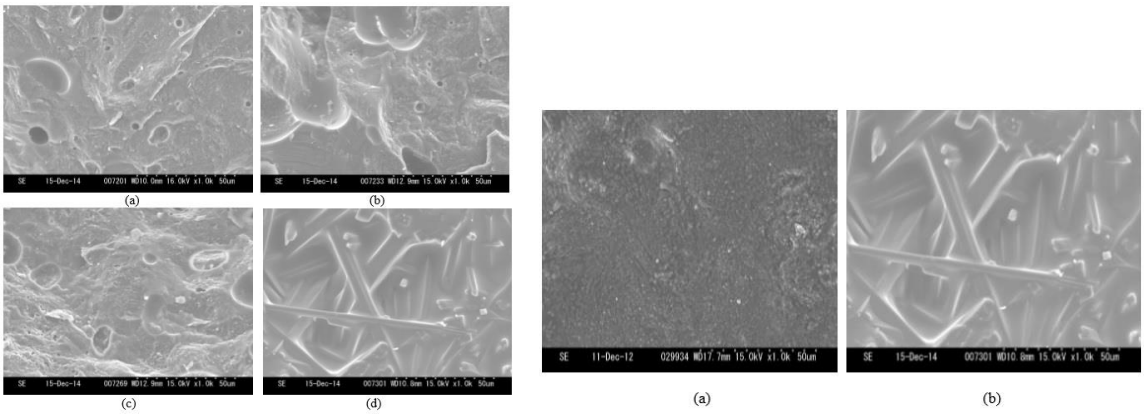
Fig. 1 (i) shows XRD patterns of silica xerogel loaded with 60 wt% α-Al_2O_3 and sintered at different temperatures. The spectrum that corresponds to a sintering temperature of 1300°C indicates a presence of alumina and mullite phases. The rate of mullite formation follows the same growth path as the sintering temperature is increased further to 1400°C and then to 1500°C. Hence, at 1600°C the complete mullite phase is formed due to the reaction of α-Al_2O_3 with the residual silica in the mixture. The mullite diffraction peaks obtained in the XRD pattern are in good agreement with other reported results [14]. Fig. 1 (ii) shows a typical X-ray diffraction pattern of samples after sintering at 1600°C by both conventional and microwave heating. It was found that in a conventional furnace, Al_2O_3 and mullite are the major phases. For samples sintered by microwave heating using a 24 GHz gyrotron, Al_2O_3 peaks were not observed and only a single homogeneous crystalline mullite phase is present. This indicates that during the sintering at 1600°C, a complete reaction of α-Al_2O_3 and xerogel silica takes place and mullite is formed. By microwave sintering, the volumetric interaction of the electromagnetic fields with the ceramic material leads to a higher heating efficiency and faster reaction rates when compared with the conventional heating at the same temperature [15].



(i) (ii)

Fig.1. XRD pattern of silica xerogel loaded by 60 wt% Al_2O_3 and sintered at (i) different temperature and (ii) 1600°C by (a) conventional heating [6], and (b) microwave heating of a 28 GHz gyrotron.

Fig. 2 (i) shows SEM images of silica xerogel loaded by 60 wt% α-Al_2O_3 and sintered at different temperatures. At a temperature of 1300°C, the microstructure is strongly heterogeneous and is characterized by the formation of secondary mullite in the form of small grains. The secondary mullite crystals are formed through a solution precipitation mechanism via transient liquid phase [16]. From 1400°C to 1500°C, it was observed a similar morphology as at 1300°C. At 1600°C, the grain shapes transform from rounded secondary to elongated primary mullite. At this temperature, the higher viscosity of the glass phase favors the growth of the crystals and stipulates the transformation of primary to secondary mullite [17]. Fig. 2 (ii) shows SEM images of fracture surfaces of the two composites, one was sintered at 1600°C in the conventional furnace and the other was sintered at 1600°C by microwave heating using a 28 GHz gyrotron. The difference in microstructure lies in the difference in grain dimension, depending on the sintering conditions. In the case of conventional sintering, small grains appear on the surface of the sample. After microwave sintering, the microstructure is characterized by a bigger grain dimension and more elongated grains. As the sintering temperature is increased (up to 1600°C), the grains growth continues.



(i) (ii)

Fig. 2. SEM images of silica xerogel loaded by 60 wt% α-Al_2O_3 and (i) sintered at (a) 1300°C, (b) 1400°C, (c) 1500°C, and (d) 1600°C, and (ii) 1600°C by (a) conventional heating [6], and (b) microwave heating of a 28 GHz gyrotron.

4. Conclusion

A solid-state transformation of a mixture of alumina and silica xerogel into mullite ceramic by microwave thermal treatment using a 28 GHz gyrotron as a radiation source has been successfully demonstrated. It has been shown that the sintering reaction using a 28 GHz gyrotron enhances significantly the grains growth and the crystallization in the silica xerogel. It was found also that a single crystalline mullite phase can be formed by microwave heating at a temperature of 1600°C for 10 min at a temperature rate of 45°C/min.

As a whole, the experimental results obtained in this study demonstrate that the microwave sintering of mullite ceramics using a 28 GHz gyrotron has clear advantages compared with the conventional treatment and can be considered as an appropriate technology for an industrial production of such materials.

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