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Synthesis and Characterization of Microwave Sintered Silica Xerogel Produced from Rice Husk Ash

I N Sudiana¹, S Mitsudo², T Nishiwaki², P E Susilowati³, L Lestari¹, M Z Firihu¹, and H Aripin⁴

¹Department of Physics, Faculty of Mathematics and Natural Science, Halu Oleo University, Kampus Bumi Tridharma Anduonohu, Kendari 93232, Indonesia
²Research Center for Development of Far-Infrared Region, University of Fukui, 3-9-1 Bunkyo, Fukui-shi 910-8507, Japan
³Department of Chemistry, Universitas Halu Oleo, Kampus Hijau Bumi Tridharma Anduonohu Kendari 93231, Indonesia
⁴Center for Material Processing and Renewable Energy, Faculty of Learning Teacher and Education Science, Siliwangi University, Jl. Siliwangi 24 Tasikmalaya 46115, West Java, Indonesia.

E-mail: muhammad.zamrun@uho.ac.id

Abstract. Silica xerogel ceramic produced from rice husk ash (RHA) taken from South East Sulawesi Indonesia has been successfully sintered by using a millimeter waves (MMW) heating system with a 28 GHz gyrotron as radiation source. The ceramic was also sintered by using an electric furnace where served as a comparison. Densification, microstructural, and morphological characterization of the silica were then investigated by using an Archimedes densification measurement method device, a X-ray diffraction (XRD) and a Scanning Electron Microscopy (SEM), respectively. Effect of microwave energy on the properties of silica xerogel ceramic were evaluated and discussed and compared to conventionally sintered results. The notably different densification and microstructure of sintered samples after sintering were found. The results suggest that microwave radiation provides a microwave effect during sintering.

1. Introduction

The silica xerogel is well recognized as one of the sintered silica materials. It is also well known that its structure shrinks considerably during drying, because the aqueous phase in the pores is removed by evaporation, capillary contraction, condensation – polymerization reactions, structural relaxation, and viscous flow [1,2]. A silica glass-ceramic is produced from this material by heat treatment process known as sintering [2,3]. The silica xerogel ceramic are extensively studied because of their high transparency and good chemical durability when exposed to irradiation. Such properties make the silica xerogel a promising candidate for applications in industry [3–5]. The properties are directly influenced by the structure of the ceramics and can be affected by several
factors, including the chemical composition, physical parameters and the characteristics of the thermal process used for production of the ceramic [6]. It is expected that an appropriately optimized thermal treatment would produce a silica glass ceramic without porosity and eventually a ceramic that has appropriate desired properties.

The conventional sintering using such as a electric furnace in silica xerogel indicates that silica glass-ceramic with a density of about 2.2 g/cm$^3$ can be obtained at a temperature as low as 980°C when compared to the conventional melting of silica glasses at about 2000°C [2]. With low heating rates, the production of this ceramic at higher sintering temperature by conventional heating needs longer time, which promotes grain growth. In contrast to the conventional (surface) heating the microwave heating is volumetric, since the electromagnetic energy is dissipated simultaneously in the whole irradiated volume. Additionally, the microwave treatment offers higher densities of the heating power, better precision in controlling the process of energy deposition etc. As radiation sources for the microwave thermal technologies both the magnetrons (with a frequency of the radiation 2.45 GHz) and the millimeter and sub-millimeter wavelength gyrotrons are considered as the most appropriate. Their application microwaves for ceramic sintering have demonstrated significant densification, improvement of the reaction rate and eventually improvement of the quality of the ceramics [7-9].

The studies on microwave treatment of alumina [7] and zirconia [8] have indicated that shorter sintering times can be achieved even at lower sintering temperatures. Moreover, in this case the samples were denser and thus stronger than the samples conventionally sintered at the same temperature for the same time duration. Additionally, the time evolution of the density, porosity, and the crystallization of silica aerogel observed in the microwave process is similar to the one observed in the conventional process [9,10], but during the microwave treatment, this evolution occurs at temperatures approximately 200°C lower. A successful use of MMW heating to sinter other materials such as boron carbide [11] and silicon nitride [12] has been reported recently. It has been found that the irradiation by millimeter waves enables very high heating rates to be achieved and reduces the time for grain growth. The smaller grains make the microstructure more uniform and increase the strength of the material. In a comparison with the magnetron heating (at frequency of 2.45 GHz) a higher power absorption and a weaker temperature dependence of the dielectric loss rate are observed when a gyrotron is used [13-15].

In this work, we present experimental results from sintering of silica xerogel extracted from rice husk ash that have been obtained on this equipment. We study and discuss the influence of the sintering temperature on the technological properties such as bulk density and microstructure of a silica xerogel ceramic. The structure of the produced silica glass-ceramic was studied using X-ray diffraction (XRD) and a Scanning Electron Microscopy (SEM).

2. Experimental procedure

2.1. Sample preparation

Silica xerogel was extracted from a rice husk ash obtained from the rice field taken from Kolaka, South East Sulawesi, Indonesia. The detailed extraction following procedures as described elsewhere [17, 18]. The rice husk was burned resulting rice husk ashes (RHA). In the pre-treatment step, RHA were washed using 1 M hydrochloric acid (HCl) at 100°C for 2 hours. Then, 10 g of RHA was mixed with 60 ml of 2 M NaOH and the mixture was boiled for 1 hour with a constant stirring. The solution was then filtered through a filter Whatman No. 41. After the filtration, the silicate was inputted to an ion-exchange resin. In the gelatin process, the sodium silicate solution was titrated with 1 N HCl under a constant stirring. The pH factor of the solution was monitored and the titration was stopped at a pH of 7 to produce a silica gel. The soft gel was aged for 18 h. After the aging, the
gel was gently stirred up by adding 100 ml of the de-ionized water to make a slurry. Washing of the aged wet gel with distilled water was done during three days, about 10 times per day. After the thorough washing the gel was dried for 12 hours at 110°C before producing a powder from it. The powder of silica xerogel was characterized by X-ray fluorescence. High purity amorphous silica amounting to 98.8% has been observed. Furthermore, the powders were ground and then polyvinyl alcohol was added as a binder. The powder was pressed at 450 MPa to form disks of 200 mm diameter and 30 mm thickness.

2.2. **Sintering experiment**

The samples were heated by using a 28 GHz gyrotron heating system in FIR Center University of Fukui, Japan with different temperatures in the interval from 300°C to 1500°C. A controlled heating rate of 45°C/min in the applicator was maintained up to the desired temperature with 10 minutes holding. The cooling was performed by a natural convection after the turning of the gyrotron off and leaving the samples inside. The sample holder was a hollow cylinder made of thermo-isolating material. The temperature profile was measured using a R-type thermocouple placed in contact with the surface of the sample. Figure 1 shows the profile of temperature as function of time of MMW sintering by using a 28 GHz gyrotron heating system.

![Figure 1. Temperature profile as function of time of MMW sintering of silica](image)

2.3. **Characterization**

After the sintering was completed, the bulk density was measured by the Archimedes's method using de-ionized water as an immersion medium. The procedure, which was followed is a standard test method described in detail by the American Society for Testing and Material Specification, ASTM C373-88 [19]. The XRD was used to determine the transition from amorphous phase to crystalline phase during the heat treatment. The intensity of the diffracted X-ray was measured as a function of the diffraction angle 2θ ranging from 0 to 90 degree. The SEM photos of fracture surface of samples were taken by using a scanning electron microscope (SEM) at a magnification of 1000-10,000 times and at accelerating voltage of 10-15 kV.
3. Experimental Results and Discussion

Figure 2 shows the relative density of samples with increased sintering temperatures in MMW (28 GHz) and conventional sintering. The 300 GHz sintering results served as a comparison is taken from reference [2]. Compared to the conventional, samples sintered by using 28 GHz showed a more rapid densification. The shift in densification varied from 0.1 to 1.0 g/cm$^3$. Furthermore, Fig. 2 also shows that densification was activated at low temperatures as low as 500°C in 28 GHz sintering. In the temperature range from 200°C to 500°C it is characterized by almost constant value for the conventional processing. This small increase in the bulk density in 28 GHz is attributed to the reduction of both the open and the closed pores. This was in contrast to the conventional heating in which at up to 900°C, the rate of sintering was nearly zero, and there was only a very small increment in density. The sample relative density at 900 °C in conventional sintering and 28 GHz was approximately 0.6 and 1.4 g/cm$^3$, respectively. It indicates a microwave effect on mass transport during sintering. These pores are ascribed to the empty sites of the evaporated water and to some residues of alcohol binder as well as to combusted residual organics when the sintering process takes place in this temperature range [1]. In the temperature range up to 900°C, the increase in bulk density is because of the condensation reactions occurs on the surface of the silanols groups (Si-OH) left in the porosity of the silica xerogel that are responsible for decreasing the open porosity. In these reactions, the silica xerogel structure tends to shrink due to the loss of the hydroxide (OH) groups. In the temperature range from 900°C to 1500°C, the silica xerogel ceramic reaches a bulk density similar to vitreous silica of about 2.2 g/cm$^3$ in a conventional system at around 980°C [2,20], while for the microwave sintering such density is approached at 900°C. A crystallization of silica glass that affects the grain growth of silica xerogel ceramic was also observed in this temperature interval. At a temperature of 800°C, crystalline cristobalite is formed from the amorphous silica. It has a lower density than the density of glass silica formed at temperatures greater than 900°C.

![Figure 2](image)

**Figure 2.** Densification of silica xerogel as a function of the sintering temperature with different sintering methods.

The increase in the density can be explained by an increase of the degree of crystallinity of cristobalite and tridymite. This crystallization affects significantly the grain growth. The sintering process provides energy, which is necessary for bonding the particles together and removing the
porosity simultaneously with the shrinking of the sizes of the particles. A moderate difference is observed comparing the bulk density of the conventional heating with that of the MMW heating in the temperature range from 500°C to 900°C. In the latter case the bulk density undergoes a progressive enhancement, while the conventional heating results in a gradual increase of this parameter. At temperatures higher than 1000°C, both the MMW and the conventional processes present a fast densification.

The effect of MMW energy on crystallization was also found. Figure 3 shows XRD patterns of the silica xerogel ceramic sintered at 800 °C by the conventional and millimeter wave processing. The XRD pattern corresponding to the conventional sintering of silica xerogel indicates an amorphous material. In contrast, the pattern for the MMW sintered silica xerogel shows the presence of a characteristic peak of cristobalite (C), indicating the beginning of the crystallization phase at about 800 °C. In literatures, it was reported that (for a conventional treatment) the silica xerogel crystallizes at temperatures between 1250 °C to 1350 °C and the process requires a long heating time (more than 800 min) in order to reach a crystalline cristobalite [20]. In the case of MMW processing, the temperature for the crystallization phase of silica xerogel was about 200 °C, lower than that observed in the conventional heating.

![Figure 3](image-url)  
**Figure 3.** XRD pattern of silica xerogel ceramic sintered at 800°C by (1) MMW and (2) conventional sintering

Figure 4 shows the SEM photographs of the fracture surfaces of microwave and conventionally sintered samples at 1000 °C. The photographs show that there are differences in particle arrangement and distribution inside the bodies. The quantitative analysis of microstructure will be performed for calculating the grain growth and will be reported elsewhere.
Figure 4. SEM silica xerogel ceramic sintered at 1000°C by (1) MMW and (2) conventional sintering

4. Conclusion

A series of experiments to investigate of microwave effects of millimeter waves sintered silica xerogel were successfully performed. A high frequency microwave sintering was performed by using a 28 GHz sintering system. The notably different behaviour of the shrinkage and microstructure evolution was found. It has been shown that the sintering reaction using millimeter wave enhances significantly the densification and the crystallization in the silica xerogel. The differences in particle arrangement and distribution inside the bodies were found from SEM photos. As a whole, the experimental results obtained in this study demonstrate that the microwave sintering of silica glass-ceramics using a millimeter wave gyrotron shows a microwave effect and has advantages compared with the conventional treatment. It can be considered as an appropriate technology for industrial production of silica ceramics.

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