



PROPERTIES OF 2.45 GHZ MICROWAVE SINTERED SiO_2 FROM RICE HUSK ASH AND Al_2O_3

Muhammad Zamrun Firihi¹ and I. Nyoman Sudiana²

¹Theoretical and Computational Physics Laboratory, Department of Physics, Faculty of Mathematics and Natural Sciences, Halu Oleo University, Kampus Hijau Bumi Tridharma Anduonohu, Kendari, Indonesia

²Material Physics Laboratory, Department of Physics, Faculty of Mathematics and Natural Sciences, Halu Oleo University, Kampus Hijau Bumi Tridharma Anduonohu, Kendari, Indonesia

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

ABSTRACT

A series of experiments of processing of two oxide ceramics (Al_2O_3 and SiO_2) as well as Al_2O_3 - SiO_2 composite by using microwave energy have been performed and reported by authors. The microwave sintering results were then compared to conventionally sintered results for every experiment to investigate a microwave effect. The reduction of processing time compared to conventional was found in all microwave experiments. In this paper, pore reduction and densification of Al_2O_3 and SiO_2 after sintering are reported. The faster pore reduction and higher in densification were observed in microwave sintered samples on both materials. The higher densification rate could be attributed to an increase in mass transport rate during sintering. The enhanced pore reduction rate in microwave sintering is promising a technology to produce a high density with fine grains of the oxide ceramics.

Keywords: oxide ceramics, microwave sintering, pore reduction, densification.

1. INTRODUCTION

The development and availability of hardware and the gradual acceptance of microwave technology in led to a remarkable growing interest on the material side. Specially on ceramic processing, application of microwave energy is increasing rapidly within the past twenty years. It is not only because it opens the possibilities for processing with low cost but also promising a new technology to get desired material properties. Generally, properties of microwave heating are: volumetric, potentially uniform in heat distribution, no thermal lag, selective heating, and quick heating. Al_2O_3 and SiO_2 are the most sintered oxide ceramics by microwaves [1]. The previous study in processing of ceramics on alumina [2-4], silica [5-6], ferrites [7-8] suggested that it is possible to produce materials by using microwave energy with fast heating. However, despite the potential implication of microwave processing for ceramics, there has been little published work which using high microwave frequency [10]. Microwave, 2.45 GHz has been the most availability and feasibility devices up to now. For ceramic processing, 2.45 GHz systems with a few kilowatts of power are also widely available. Recently, an advanced progress in the development of microwaves sources has been produced a material processing material system with higher frequency electromagnetic waves, such as, millimeter and submillimeter waves [11-14]. However, these devices are still limited available because very expensive for ordinary laboratory.

Abundance and availability of agricultural wastes can be processing to be a more valuable product. Agricultural by product are usually inexpensive, and their effective utilization is a desired outcome. Rice husk is one of agriculture wastes which abundant and not utilized properly for significant industrial use in Indonesia except to a small extent as animal seeds supplement. It was found that when rice husks are burnt, the resulting back

ash contains over than 60 % of silica. The silica of rice husks with many impurities can be converted into high purity amorphous silica. Other application of rice husk has been reported by Nornikman, *et al.*, for microwave absorber [15].

In this work, silica ceramics from local rice husk as well as a commercial alumina powder were prepared and sintered by using a microwave of 2.45 GHz. Examination of pore reduction and density of material after sintering was performed. Results of conventional sintering were used as comparison.

2. MATERIAL AND METHOD

Silica powder was produced from rice husk ash from Kendari, Indonesia. The procedure of extracting silica from rice husk ash has been published elsewhere [16]. The starting powder of alumina was used a commercial alumina powder, an AES-11C. The powders were dispersed in de-ionized water with 0.3wt% dispersant, an ammonium polycarboxylate acid to form slurry. The dispersant was added to the slurry to prevent the agglomeration of the suspended powder particles. Cylindrical samples were then formed. After formed, the samples were pressed and followed by pre-sintered. After pre-sintering, alumina compacts were then sintered by using microwaves and conventional. A 2.45 GHz microwave oven was applied for microwave heating system. The temperature was measured by using thermocouple equipped with a controlling system. In this system, the temperature was measured carefully, and has been tested in several experiments. In order to achieve the homogenous heating, the sample was placed inside a alumina fiber-board as thermal insulation. Because of the use of thermal insulation, we can assume that no significant temperature difference between the surface and internal sample occurred in experiments. After holding the samples at the desired temperature, cooling was carried



out naturally with the samples left inside the oven. The conventional sintering was performed by using an electric furnace. The sintering temperatures were from a temperature of 400 °C to 1200 °C in air. Density of the samples was measured based on the Archimedes's method where de-ionized water used as an immersion medium. The procedure used in this study was concomitant with the standard test method described in detail by the American Society for Testing and Material Specification, ASTM C373-88 [18]. Porosities were calculated from SEM photos. Before take a SEM, sample surface was coated with evaporated platinum (Pt) before SEM photos were taken. Gray scale SEM photos were processed by various images processing programs before the pore calculation.

The total pore was then determined for each sintering temperature by Image software.

3. RESULTS AND DISCUSSIONS

3.1 Sintering of alumina

Figure-1 shows pore reduction of alumina after sintering by microwave (2.45 GHz) and conventional (electric furnace). It shows a faster pore reduction in microwave sintering. Moreover, pore reduction is also start at earlier sintering temperature in the microwave sintering method. It is indicated that different sintering kinetic between two sintering process.

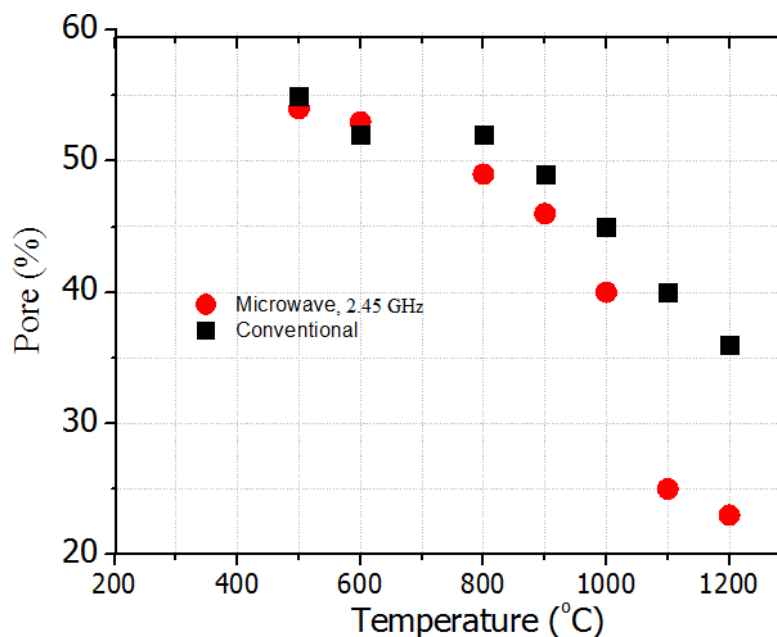


Figure-1. Pore reduction of alumina with increasing sintering temperature.

Figure-2 shows densification of sintered alumina with increase in sintering temperature for alumina. Compared to the conventional sintering, samples sintered in microwave showed a more rapid densification. The greatest shift in densification is approximately 8 % at

sintering temperature 1200°C. Such phenomena on densification of alumina has been also reported previously [9]. The densification result agrees with the porosity reduction result shown in Figure-1.

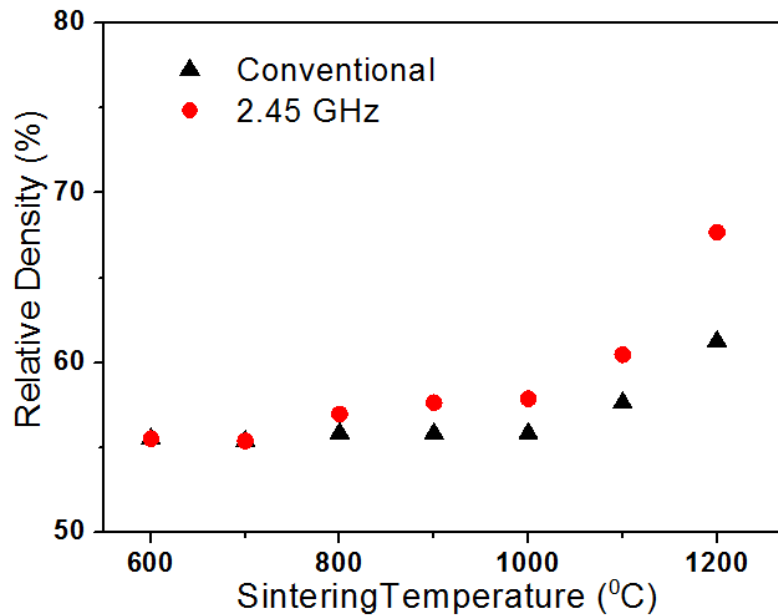


Figure-2. Densification profile of microwave and conventionally sintered alumina.

3.2 Sintering of silica

Experiments of sintering silica from rice husk ash by using microwave energies were successfully performed by using 2.45 GHz. Some experiment results are presented in this section. Figure-3 shows the reduction of porosity of

silica samples with increased sintering temperatures in microwave and conventional sintering. Compared to the conventional, samples sintered by using microwave indicating a more efficient removal of porosity.

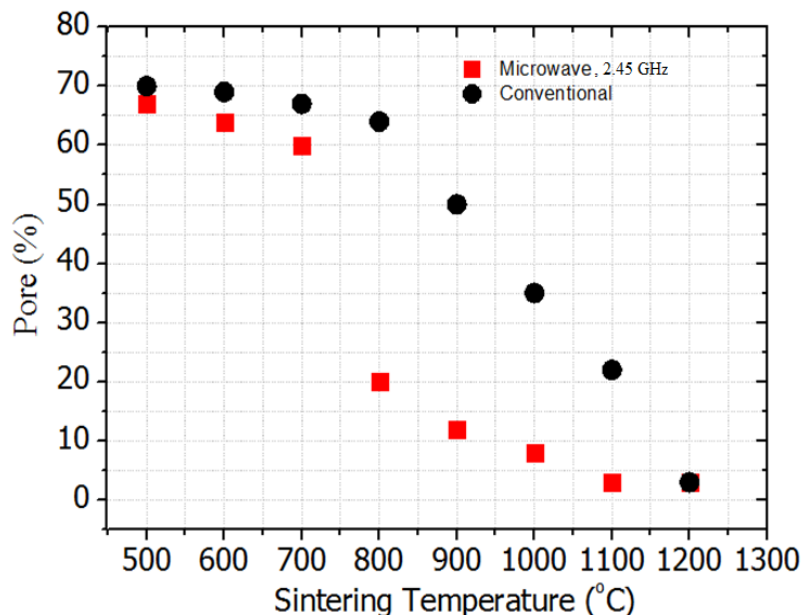


Figure-3. Pore reduction of silica with increasing sintering temperature.

At low temperature below 800°C, 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 [14]. In the high temperature

of 900 to 1200°C, the increase in density is because of the condensation reactions occurs on the surface of Si-O-H left in pores of the silica that are responsible for decreasing the porosity.



A comparison of Figure-1 and Figure-3 shows that the difference in the amount of porosity between microwave and conventional is more pronounced in silica than in alumina. For silica, in the temperature range up to 800°C it is characterized by almost constant porosity for the conventional processing. However, these results suggested that microwave fields enhance pore reduction

on both ceramics. Figure-4 shows the densification behaviour of silica after sintered by using microwave and electric furnace (conventional). As in sintering of alumina, the graph shows a faster densification on microwave sintering. The result suggests that microwave enhances sintering during silica processing.

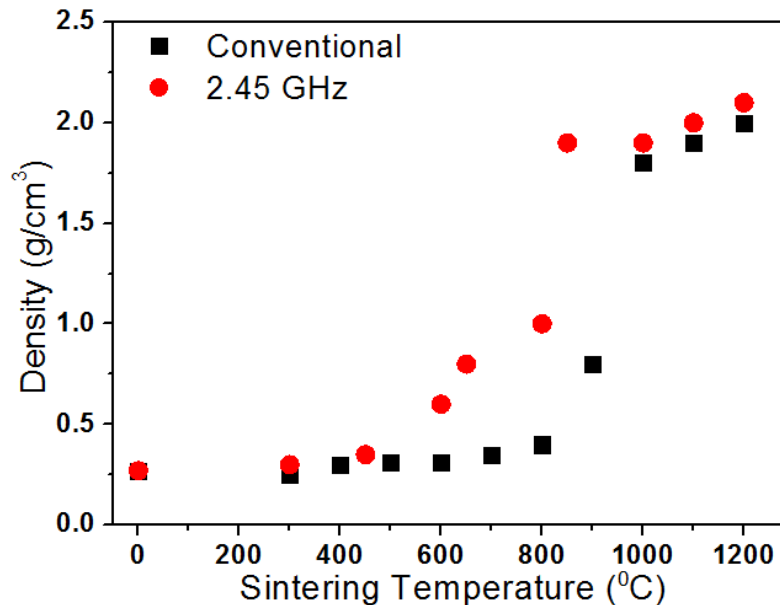


Figure-4. Densification profile of microwave and conventionally sintered of silica.

The faster pore reduction and higher in densification observed in microwave sintered oxide ceramics could be associated with atom diffusion rate during sintering [19]. This could be associated to the increase in either driving force or apparent activation energy of diffusion. The preliminary estimation of apparent activation energy of alumina revealed that the apparent activation energy of microwave sintering is much lower than that in conventional sintering samples. The details of experiment results of apparent activation energy estimation will be published elsewhere in separate paper. Next experiment will be prepared for more details characterization of sintered samples to reveal more microwave mechanism of microwave materials.

4. CONCLUSIONS

Processing oxide ceramics by using microwave energy, 2.45 GHz, to produce a high density compact was successfully performed. Two powders were applied as starting materials. Compared to conventional results, the densification of microwave at same sintering temperature is much higher. The results found in this study suggest that microwaves increase in atom diffusion rate during sintering.

ACKNOWLEDGMENTS

This work has been partially supported by the Directorate of Research Strengthening and Development,

at the Ministry of Research, Technology and Higher Education, Republic of Indonesia

REFERENCES

- [1] W. H. Sutton. 2005. Microwave processing of Ceramic Materials. Microwave Solutions for Ceramic Engineers, American Ceramic Society. 35-65.
- [2] K. H. Brosnan, G. L. Messing, and D. K. Agrawal. 2003. Microwave Sintering of Alumina at 2.45 GHz, J. of the Am. Cer. Soc. 86(8): 1307-1312.
- [3] I. N. Sudiana and M. Z. Firihi. 2016. Effect of initial green samples on mechanical properties of alumina ceramic, Contemporary Engineering Sciences. 9(12): 595-602.
- [4] I. N. Sudiana, S. Mitsudo, M. Z. Firihi, and H. Aripin. 2016. Effect of High-Frequency Microwave on Micro Hardness of Alumina Ceramic, Material Science Forum. 872: 114-117.
- [5] H. Aripin, S. Mitsudo, E. S. Prima, I. N. Sudiana, S. Tani, K. Sako, Y. Fujii, T. Saito, T. Idehara, S. Sano, B. Sunendar, and S. Sabchevski. 2012. Structural and



- Microwave Properties of Silica Xerogel Glass-Ceramic Sintered by Sub-millimeter Wave Heating using a Gyrotron, *J. Infrared, Millimeter, and Terahertz Waves*. 33(11): 1149-1162.
- [6] H. Aripin, S. Mitsudo, E. S. Prima, I. N. Sudiana, H. Kikuchi, Y. Fujii, T. Saito, T. Idehara, S. Sano, and S. Sabchevski. 2015. Crystalline mullite formation from mixtures silica xerogel converted from sago-of alumina and a novel material waste ash, *Ceramics International*. 41(5): 6488-6497.
- [7] P. Yadoji, R. Pamedu, D. Agrawal, and R. Roy. 2003. Microwave sintering of Ni-Zn ferrites: comparison with conventional sintering, *Materials Science and Engineering: B*. 98(3): 269-278.
- [8] I. N. Sudiana, S. Mitsudo, T. Nishiwaki, P. E. Susilowati, L. Lestari, M. Z. Firihi, and H. Aripin. 2015. Effect of Microwave Radiation on the Properties of Sintered Oxide Ceramics, *Contemporary Engineering Sciences*. 8(34): 1607-1615.
- [9] M. Z. Firihi and I. N. Sudiana. 2016. Microwaves Enhanced Sintering Mechanisms in Alumina Ceramic Sintering Experiments. *Contemporary Engineering Sciences*. 9(5): 237-247.
- [10] I. N. Sudiana, R. Ito, S. Inagaki, K. Kuwayama, K. Sako, and S. Mitsudo. 2013. Densification of Alumina Ceramics Sintered by Using Sub-millimeter Wave Gyrotron, *J. Infrared, Millimeter, and Terahertz Waves*. 34(10): 627-638.
- [11] H. Aripin, S. Mitsudo, E. S. Prima, I. N. Sudiana, H. Kikuchi, S. Sano, and S. Sabchevski. 2013. Microstructural and Thermal Properties of Nanocrystalline Silica Xerogel Powders converted from Sago Waste Ash Material, *Materials Science Forum*. 737: 110-118.
- [12] H. Aripin, S. Mitsudo, I. N. Sudiana, T. Saito, and S. Sabchevski. 2015. Structure Formation of a Double Sintered Nanocrystalline Silica Xerogel Converted From Sago Waste Ash, *Transactions of the Indian Ceramic Society*. 74(1): 11-15.
- [13] S. Mitsudo, H. Hoshizuki, T. Idehara, and T. Saito. 2006. Development of material processing system by using a 300 GHz CW gyrotron, *J. Phys.: Conference Series*. 52: 549-552.
- [14] S. Mitsudo, K. Sako, S. Tani, and I. N. Sudiana. 2011. High Power Pulsed Submillimeter Wave Sintering of Zirconia Ceramics, *The 36th Int. Conf. on Infrared, Millimeter and THz Waves (IRMMW-THz 2011)*, Hyatt Regency Houston, Houston, Texas, USA.
- [15] H. Nornikman, F. Malek, L. Y. Seng, M. H. Ramli, N. A. M. Syafiq, M. H. Mazlan, M. Z. A. Abd Aziz, B. H. Ahmad and A. Salleh. 2015. Green Technology Design of Modified Wedge Microwave Absorber Using Rice Husk. *ARNP Journal of Engineering and Applied Sciences*. 10(17): 7380-7385.
- [16] H. Aripin, S. Mitsudo, I. N. Sudiana, S. Tani, K. Sako, Y. Fujii, T. Saito, and T. Idehara. 2011. Rapid Sintering of Silica Xerogel Ceramic derived from Sago Waste Ash Using Submillimeter Wave Heating of a 300 GHz CW Gyrotron, *J. Infrared and Millimeter Waves*. 32(6): 867-876.
- [17] S. Mitsudo, S. Inagaki, I. N. Sudiana, and K. Kuwayama. 2013. Grain Growth in Millimeter Wave Sintered Alumina Ceramics, *Advanced Materials Research*. 789: 279-282.
- [18] ASTM Standard C373 - 88. 2006. Standard test method for water absorption, bulk density, apparent porosity and apparent specific gravity of fired whiteware products, *ASTM International*, West Conshohocken, PA.
- [19] M. A. Janney and H. D. Kimrey. 1990. Microwave Processing of Materials II, *Materials Research Society Proceeding*. 189: 215-228.