Contemporary Engineering Sciences, Vol. 8, 2015, no. 34, 1607 - 1615
HIKARI Ltd, www.m-hikari.com
http://dx.doi.org/10.12988/ces.2015.511303

Effect of Microwave Radiation on the Properties of Sintered Oxide Ceramics

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Abstract

Recent research has been significantly increased our fundamental understanding of microwave interactions with materials. Thermal absorption has been demonstrated to result from simultaneous action of multiple dissipation mechanisms during processing. In addition, it has been conclusively established that strong microwave fields exert a non-thermal driving force during sintering. This force acts as an additional driving force for atomic transport. For strong electric fields, the force can enhance diffusion rates during ceramic sintering. This paper describes recent research on microwave sintering of two oxide ceramics, a silica xerogel ceramic produced from rice husk ash (RHA) and a high purity alpha alumina. A millimeter waves (MMW) heating system with a 28 GHz gyrotron is applied for microwave sintering experiment. The ceramics were also sintered by using an electric furnace where served as comparison. Effect of microwave energy on the porosity reduction of the ceramics was investigated. Some possible physical mechanisms were discussed.

Keywords: Sintering, microwave, microwave effect, silica xerogel, alumina

1 Introduction

Silica (SiO_2) and alumina (Al_2O_3) are well recognized as the most sintered oxide materials. Silica 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]. The silica xerogel ceramic are also extensively sintered because of their potential application in industry [1–5]. Microwave sintering can be one of appropriate candidate processing methods to get high quality silica ceramics.

Alumina are the most microwave sintered material because their simple sintering mechanism. So that mechanism of microwave interaction with the material may be more clearly understood. The previous reports in microwave sintering of alumina ceramic suggested that the microwave enhances densification of alumina. It also depend on microwave frequencies [6,7]. It suggested that it is possible to produce a higher density alumina compact by using a higher microwave frequency. That is useful for alumina application.

The conventional sintering using such as a electric furnace on ceramics indicates that silica glass-ceramic with a density of about 2.2 g/cm3 can be obtained at a temperature as low as 980°C when compared to the conventional melting of silica glasses at about 2000°C [1]. For alumina the highest density can be found at temperature1700°C [6,7]. By using traditional or conventional sintering method needs longer time, which not only requires a lot of energy consumption but also promotes grain growth. In contrast to the conventional heating the microwave heating is volumetric, since the electromagnetic energy is dissipated simultaneously in the whole irradiated volume. So that need shorter processing time. Microwave radiation sources available up to now are magnetrons for microwave frequency of 2.45 GHz and gyrotrons for higher frequencies such as millimeter and sub-millimeter waves. The application gyrotrons for material processing system have been reported for several functional ceramics. Some unexpected results indicated as microwave effects were found [6-10]. Comparing to processing by using 2.45 GHz, a higher power absorption and a weaker temperature dependence of the dielectric loss rate are observed at higher microwave frequencies [1, 11-14].

In this work, we present investigation results on microwave sintering of silica xerogel and high purity alpha alumina that have been obtained by using the MMW sintering system. Microwave effects on ceramic properties were also addressed.

2 Experimental Setup

2.1 Sample preparation

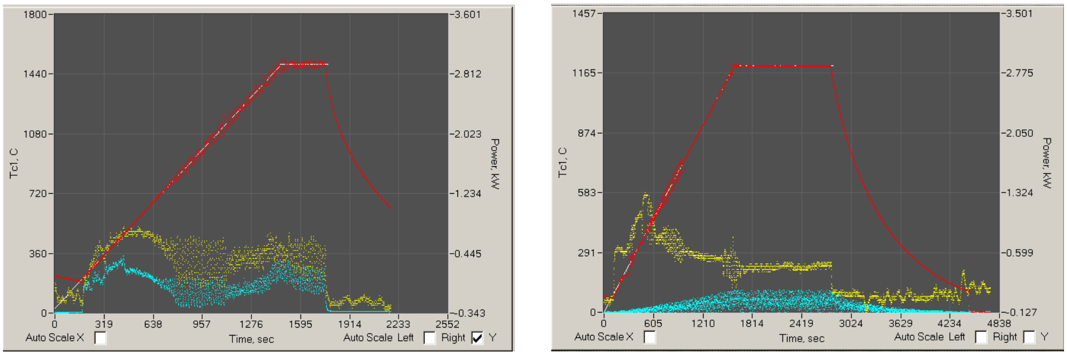
An high purity alpha alumina powder AES-11C (99.8 %) and silica xerogel extracted from a rice husk ash obtained from the rice field taken from Kolaka, South East Sulawesi, Indonesia were used as starting material. The rice husk was burned resulting rice husk ashes (RHA). The detailed extraction following procedures as described elsewhere [15]. Samples were prepared by pressed the powder to form disks before sintering.

2.2 Sintering Experiment

The samples were processed by using a millimeter wave (28 GHz) gyrotron heating system at FIR Center University of Fukui, Japan with sintering temperatures up to 1700°C. A controlled heating rate of 45°C/min in the applicator was maintained up to the desired temperature. The sample holder was made of heat insulations material (Fibermax). The temperature 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 for both materials. The figures show difference microwave power consumption to be increasing the temperatures of silica and alumina upon sintering.

2.3 Characterization

After the sintering was completed, the bulk density, open porosity, and close porosity were measured by the Archimedes 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 [17]. The SEM photos of samples were taken by using a scanning electron microscope (SEM). Properties of microwave sintered ceramics were then compared to conventionally sintered ones.



Microwave Power (kW) Sample's Temperature
Chamber's Temperature

Figure 1. Temperature profile as function of time of MMW sintering of silica and alumina ceramics

3 Microwave Effects Analysis

Figure 2 shows the reduction of open and closed porosity of silica samples with increased sintering temperatures in MMW (28 GHz) and conventional sintering. Compared to the conventional, samples sintered by using MMW indicating a more efficient removal of closed porosity. Figure 3 shows the reduction of open and closed porosity of alumina samples. The densification of alumina has been reported previously [7]. A comparison of Figure 2 and Figure 3 shows that the difference in the amount of closed porosity between MMW and conventional is more pronounced in silica than in alumina. For silica, in the temperature range from 200°C to 900°C it is characterized by almost constant porosity for the conventional processing. Reduction on both open and closed pores is shown in MMW. 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-O-H) left in the porosity of the silica xerogel that are responsible for decreasing the open porosity [1,16]. In contrast to the conventional, MMW shows rapid open pore reduction starting from temperature 600°C as shown in Fig. 2. For alumina, the open pore reduction in MMW start at 900°C and conventional start at 1100°C as shown in Fig. 3. It suggested that microwave fields enhance pore reduction on both ceramics.

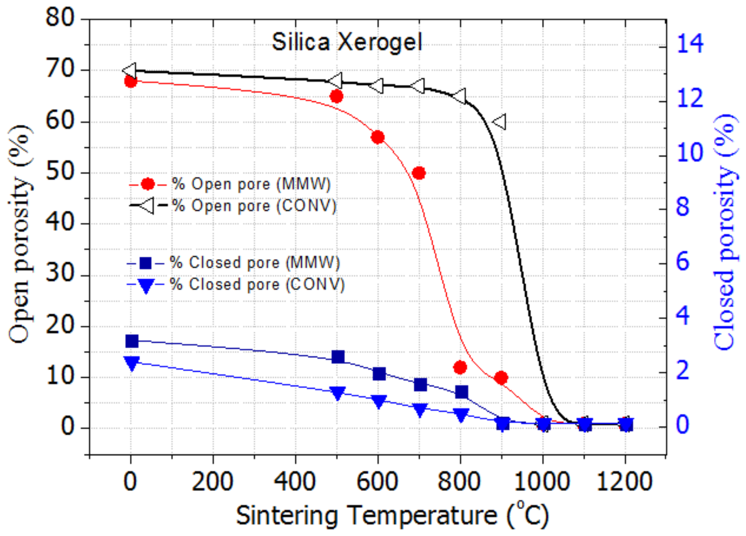


Fig. 2 Reduction of open and closed porosity of silica xerogel upon MMW as compared to conventional sintering

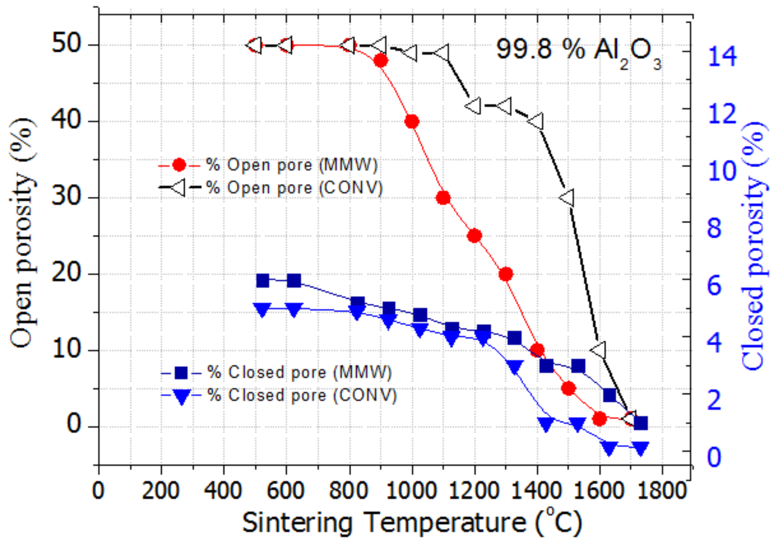


Fig. 3 Reduction of open and closed porosity of alumina upon MMW as compared to conventional sintering

In addition, effect of microwaves on crystallization of silica was also reported [18]. 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 4 shows the SEM photographs of the surfaces of microwave and conventionally sintered alumina at 1400 °C. The photographs show that there are differences in pore distribution inside the bodies. The detail quantitative analysis of pore structure will be performed and reported elsewhere. The observations related to porosity can be analyzed using classical sintering theory, which defines the local driving force for pore removal and densification by the energy balanced characterized [19]. The dielectric inhomogeneity of a porous materials and the pore shape is assumed to cause and electric field strength enhancement at surface and provide a driving force for pore removal.

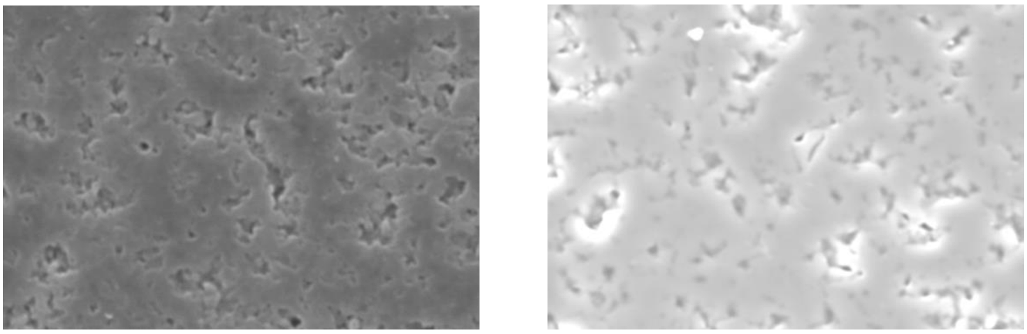


Figure 4. SEM photos of porosity of alumina ceramics sintered at 1400^{o}C by (1) MMW (left) and (2) conventional sintering (right)

4 Conclusion

Microwave effects based on porosity changes upon MMW sintering of two oxide ceramics are reported. The MMW shows faster removal porosity than conventional. It can be applied to enhance densification and to reduce the grain growth. The differences in porosity inside the bodies were found from SEM photos. In order to explain this effect, the existing sintering model based on thermodynamic stability of pores surrounded by grains can be applied for the case of an external field. It suggests that the microwave field would provide an additional driving force for pore removal. This study demonstrate that the microwave sintering by using MMW shows a microwave effect and has advantages compared with the conventional and can be considered as an appropriate technology for processing oxide ceramics.

Acknowledgements. This work has been partially supported by the Directorate of Higher Education, at the Ministry of Research Technology and Higher Education, Republic of Indonesia and FIR Center, University of Fukui, Japan.

References

[1] H. Aripin, S. Mitsudo, I. N. Sudiana, S. Tani, K.Sako, Y. Fujii, T. Saito, Toshitaka Idehara, Rapid Sintering of Silica Xerogel Ceramic derived from Sago Waste Ash Using Submillimeter Wave Heating of a 300 GHz CW Gyrotron, *Journal of Infrared, Millimeter, and Terahertz Waves*, **32** (2011), 867-876. http://dx.doi.org/10.1007/s10762-011-9797-2

[2] H. El Hamzaoui, L. Courtheoux, V. N. Nguyen, E. Berrier, A. Favre, L. Bigot, M. Bouazaoui, B. Capoen, From porous silica to bulk optical glasses: The control of densification, *Mater. Chem. And Phy*., **121** (2010), 83 – 88. http://dx.doi.org/10.1016/j.matchemphys.2009.12.043

[3] N.A. Melosh, B.J. Scott, C. Steinbeck, R.C. Hayward, P. Davidson, G.D. Stucky, and B.F. Chmelka, Mesostructured Silica/Block Copolymer Composites as Hosts for Optically Limiting Tetraphenylporphyrin Dye Molecules, *J. Phys. Chem. B*, **108** (2004), 11909–11914. http://dx.doi.org/10.1021/jp040064m

[4] H. Xiu, Y. Pan, H. Kou, Y. Zhu, J. Guo, Preparation of transparent ordered mesoporous carbon/silica composite and their optical limiting properties, *J. of Alloy and Comp*., **502** (2010), L6 – L9. http://dx.doi.org/10.1016/j.jallcom.2010.03.173

[5] Y.T Kim, D.S Kim, D.H. Yoon, PECVD SiO2 and SiON films dependent on the rf bias power for low loss silica waveguide, *Thin Solid Film*, **475** (2005), 271 – 274. http://dx.doi.org/10.1016/j.tsf.2004.07.044

[6] I. N. Sudiana, R. Ito, S. Inagaki, K. Kuwayama, K. Sako, S. Mitsudo, Densification of Alumina Ceramics Sintered by Using Sub-millimeter Wave Gyrotron, *Int. J. of Infrared, Millimeter, and Terahertz Waves*, **34** (2013), 627-638. http://dx.doi.org/10.1007/s10762-013-0011-6

[7] M.A. Janney, H.D. Kimrey, Microwave Processing of Materials II, *Materials Research Society Proceeding*, **189** (1990), 215-228.

[8] S. Mitsudo, K. Sako, S. Tani, I.N. Sudiana, High Power Pulsed Submillimeter Wave Sintering of Zirconia Ceramics, *The 36th Int. Conference on Infrared, Millimeter and THz Waves (IRMMW-THz 2011)*, Hyatt Regency Houston, Houston, Texas, USA, 2011, October 2-7.

[9] H. Aripin, S. Mitsudo, E.S. Prima, I.N. Sudiana, H. Kikuchi, Y. Fujii, T. Saito, T. Idehara, S. Sano, S. Sabchevski, Crystalline mullite formation from mixtures of alumina and a novel material - Silica xerogel converted from sago waste ash, *Ceramics International*, **41** (2015), 6488–6497.

[10] S. Mitsudo, H. Hoshizuki, K. Matsuura, T. Saji, T. Idehara, M. Y Glyavin, A. G. Eremeev, T. Honda, Y.Iwai, A. Kitano, J. Ishibashi, H. Nishi, Ultra high temperature sintering by using 24 GHz gyrotron, *Proc. of 15-th Int. Conf. on High Power Particle Beams*, Saint-Petersburg, Rusia, (2010), 528 – 531.

[11] S. Mitsudo, R. Ito, I.N. Sudiana, K. Sako, and K. Kuwayama, *Grain Growth*

in Submillimeter Waves Sintered Alumina, IRMMW-THz, Wollongong, Australia, September, 2012.
http://dx.doi.org/10.1109/msmw.2004.1346173

[12] Y. V Bykov, O. I Get'man, V. V Panichkina, I. V Plotnikov, V. V Skorokhod, V. V Kholoptsev, Sintering of Si3N4 ceramics with additives containing yttrium and ytterbium oxides with microwave and conventinal heating, *Powder Metallurgy and Metal Ceramic*, **40** (2001), 112 – 120.
http://dx.doi.org/10.1023/a:1011915103130

[13] H. Aripin, S. Mitsudo, E. S. Prima, I. N. Sudiana, S. Tani, K. Sako, Y.Fujii, T. Saito, T. Idehara, S. Sano, B. Sunendar, S. Sabchevski, Structural and Microwave Properties of Silica Xerogel Glass-Ceramic Sintered by Sub-millimeter Wave Heating using a Gyrotron, *Int. J. of Infrared, Millimeter, and Terahertz Waves*. **33** (2012), 1149-1162.
http://dx.doi.org/10.1007/s10762-012-9925-7

[14] H. Aripin, S. Mitsudo, I.N. Sudiana, T. Saito, S. Sabchevski, Structure Formation of a Double Sintered Nanocrystalline Silica Xerogel Converted From Sago Waste Ash, *J. Transaction of Indian Ceramics Society*, **74** (2015), 11-15. http://dx.doi.org/10.1080/0371750x.2014.980850

[15] S. Affandi, H. Setyawan, S. Winardi, A. Purwanto, R. Balgis, A facile method for production of high purity silica xerogels from bagasse ash, *Advance Powder Technology*, **20** (2009), 468 – 472.
http://dx.doi.org/10.1016/j.apt.2009.03.008

[16] C. Folgar, D. Folz, C. Suchicital, D. Clark, Microstructure evolution in silica aerogel, *J. of Non-Cryst. Solid*, **353** (2007), 1483 – 1490.
http://dx.doi.org/10.1016/j.jnoncrysol.2007.02.047

[17] ASTM, Directory of testing laboratories, commercial-institutional/compiled by the American Society for Testing Material, *ASTM: Philadelphia*, (1975), C373 – C388.

[18] H. Aripin, S. Mitsudo, E. S. Prima, I. N. Sudiana, H. Kikuchi, S. Sano, S. Sabchevski, Microstructural and Thermal Properties of Nanocrystalline Silica Xerogel Powders converted from Sago Waste Ash Material, *Material Science Forum*, **737** (2013), 110-118.
http://dx.doi.org/10.4028/www.scientific.net/msf.737.110

[19] B. J. Kellett and F. F. Lange, Thermodynamics of Densification: I, Sintering of Simple Particle Arrays, Equilibrium Configurations, Pore Stability, and Shrinkage, *J. of American Ceramic Society*, **72** (1989), no. 5, 725–734.
http://dx.doi.org/10.1111/j.1151-2916.1989.tb06208.x

Received: October 1, 2015; Published: December 5, 2015