

A Comparative Study of Double Layers $\text{Al}_2\text{O}_3/\text{Al}_2\text{O}_3$ and $\text{Al}_2\text{O}_3/\text{SiO}_2$ Prepared By Microwave and Natural Drying

N.F. Ismail*, Z.Harun, N.A.Badarulzaman

Faculty of Mechanical and Manufacturing Engineering, Department of Material and Design Engineering, Universiti Tun Hussein Onn Malaysia, 86400 Parit Raja, Batu Pahat, Johor, Malaysia.

Received 22 June 2011; accepted 25 May 2012, available online 13 Sep 2012.

Abstract: In this work, a comparative study of green double layers ceramic bodies prepared by natural and microwave drying techniques was investigated. The double layers ceramic with combination of $\text{Al}_2\text{O}_3/\text{Al}_2\text{O}_3$ and $\text{Al}_2\text{O}_3/\text{SiO}_2$ samples were prepared via compaction method. Alumina powder, Al_2O_3 with particle size $90\mu\text{m}$ and $<45\mu\text{m}$, silica powder, SiO_2 with particle size $<45\mu\text{m}$ were used in this study. Measurement of linear shrinkage of pellet shows that microwave drying contributed to lower shrinkage value compared to the natural drying method. The characterization of the microstructure of double layers ceramic body also showed that microwave technique formed a good bonding structure especially at the critical interface layer. In fact, the implementation of microwave drying also significantly reduced the drying time and produced a good and homogenous final structure.

Keywords: Ceramic pellet, microwave drying, microstructure, linear shrinkage

1. Introduction

Most of the ceramic component that used in varies application *i.e* membrane, mould for casting and wall consists of multilayered structure [1,2]. Basically, ceramic double layers structure is fabricated with two similar and dissimilar materials and these layers can be connected by the diffusion bonding [3]. Diffusion bonding of layered structures can be established via solid state or liquid state sintering process. This sintering stage is needed in order to connect the two similar and dissimilar materials. During diffusion stage several physical and metallurgical processes occur simultaneously, such as creep, diffusion, dislocation formation, recrystallization and formation of reaction product at the interface. Typically, bonding of the ceramic interfaces depending on the materials used such as alumina, silica, and titania.

Drying is one of the most important processes and complicated process for ceramic materials due to the fact that ceramic body will typically shrink during firing [4-7]. This shrinkage phenomenon can lead to cracking mechanism and loss the quality of the final products. A crucial task in the drying process of ceramic green bodies is the minimization of non uniform drying stresses in order to prevent cracking and non uniform deformation in the green body structure [8]. Drying can be defined as a process of removing water from unfired body or raw materials in the green body. Drying will be accomplished by supplying energy to the ceramic in order to complete evaporation and drying by evaporation is commonly applied to a large number of materials for instance ceramic [9, 15]. Drying process can change the microstructure of materials which consequently affect the characteristic of the dried materials. Some of the

materials must be dried slowly and carefully to avoid crack, some have to be dried as thoroughly as possible and others need very low temperature [16]. Several methods have been utilized for the purpose of ceramic drying starting with conventional drying method such as natural air drying and oven drying. However, these methods are not always sufficient. Over the past years, the microwave drying method is becoming more and more important regarding to advance or faster drying [10]. Besides, the microwave processing of materials offers the potential of reducing production time of ceramic materials and this advantage of speed up of drying process can lead up the reduction of energy requirement in processing [11]. Basically, the conventional drying is slow in thick materials and can cause the drying process taking the longer time [12]. The natural drying is not only slow but nonuniform where the surface is hotter than inside. In the previous studies [13, 14], the microwave drying has been proved that this technique offered the advantage especially in drying acceleration and during microwave drying, the samples is heated from inside to the outside [17]. Thus, this technique is needed for the thick materials especially for double layers structure due to the microwave can penetrate internally and the interior part become hotter than its surface. This energy can heat materials faster than other drying method and the microwave drying offered possibility of predicting higher reduction in drying time.

In this study, the double layers ceramic pellet were fabricated from $\text{Al}_2\text{O}_3/\text{Al}_2\text{O}_3$ and $\text{Al}_2\text{O}_3/\text{SiO}_2$ using powder compaction technique and then dried with different drying modes; microwave and natural. Double layers of $\text{Al}_2\text{O}_3/\text{Al}_2\text{O}_3$ and $\text{Al}_2\text{O}_3/\text{SiO}_2$ ceramics are expected to be used in a wide range application, such as for filters or membrane for gases and liquids,

*Corresponding author: far_hani11@yahoo.com
2012 UTHM Publisher. All right reserved.
penerbit.uthm.edu.my/ojs/index.php/ijie

semiconductor industries and structural products [18]. Basically, ceramic membranes consisting of several layers of one or more different ceramic materials [19]. The aim of this study was to investigate the influence of drying technique to the microstructure of the ceramic layer and properties. The results presented provide a basic understanding the effect of drying on the double layers ceramic bonding sintered structure.

2. Experimental procedure

2.1 Sample Preparation

A commercial alumina powder (purity of 99%, Sigma Aldrich) and silica (industrial grade) were used in the preparation of double layers ceramic. Then, the alumina and silica powder with different particle size were added with 5% binder, polyvinyl alcohol (PVA) and the mixture was milled for 30 minutes by using ball mill until the mixture was homogenized. The two groups of double layers pellets $\text{Al}_2\text{O}_3/\text{Al}_2\text{O}_3$ and $\text{Al}_2\text{O}_3/\text{SiO}_2$ were fabricated with two similar and dissimilar materials see Fig.1. The particle sizes of $90\mu\text{m}$ and below $45\mu\text{m}$ were used for double layers $\text{Al}_2\text{O}_3/\text{Al}_2\text{O}_3$ structure whereas alumina ($90\mu\text{m}$) and silica (below $45\mu\text{m}$) were used for $\text{Al}_2\text{O}_3/\text{SiO}_2$ pellets. The pressure applied on the samples was 3 tons for 3 minutes for each of the layer.

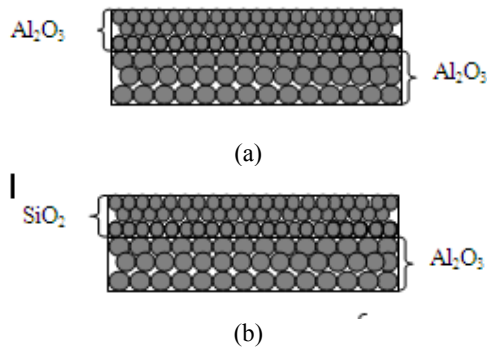


Fig. 1 (a) and (b) Schematic representation of ceramic double layered structures

2.2 Drying Conditions

After the desired double layers samples were fabricated, the samples were dried by using microwave and natural drying. Under microwave drying, the samples were removed periodically from the microwave for every 10 minutes. This procedure was repeated until the desired moisture content (constant in weight) was achieved. The time taken for the samples to reach desired moisture content also was recorded. On the other hand, natural drying also was carried out at room temperature for 24 hours.

After drying process, the green bodies were sintered in a temperature programmable furnace. The sintering involves two stages, the first stage is 500°C in order to burn out the binder, PVA and followed by densification of ceramic bodies at 1550°C , both with the heating rates

of $2^\circ\text{C}/\text{min}$. The samples were soaked for 3 hours during the densification process.

2.3 Properties Characterizations

The sintered double layers ceramic structures were characterized by physical properties and microstructural analysis. The linear shrinkage of double layered structures was determined using the following equations:

$$\text{Shrinkage} = \frac{d_i - d_f}{d_i} \times 100\% \quad (1)$$

where d_i is the diameter of sample before drying and d_f is the diameter after drying of green and sintered double layered structure. The diameter was measured by vernier caliper (Mitutoyo) and average values were taken.

Archimedes' method was used for calculation of the density and porosity of the sample after sintering process. The measurement of density and porosity were carried out by using Mettler Toledo Density Kit. The density and porosity of the ceramic pellets were calculated by using mass of dried sample, W_d , mass of immersed and suspended specimen in liquid, W_s and mass of immersed and suspended specimen in air, W_w .

$$\text{Porosity} = \frac{W_w - W_d}{W_w - W_s} \times 100\% \quad (2)$$

$$\text{Density} = \frac{W_d}{W_d - W_s} \times 100\% \quad (3)$$

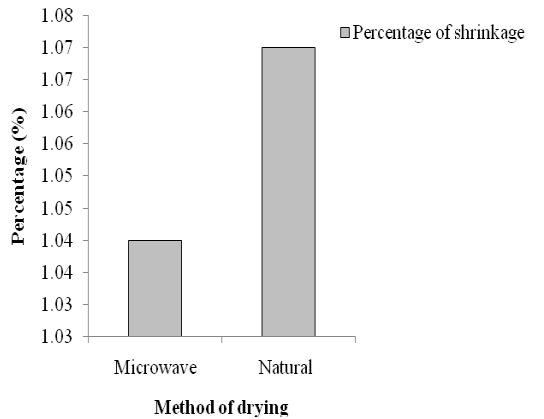
Scanning electron microscope, SEM (JEOL, JSM-6380LA) was used to examine the cross section of the sample. The sample micrographs were observed by using back scattered electron mode using $100\times$ magnifications.

3. Experimental procedure

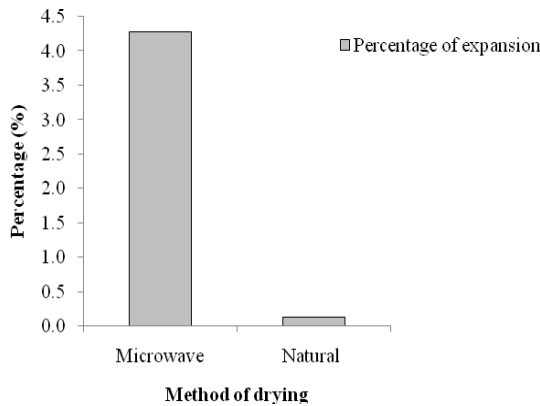
3.1 Linear Shrinkage

Shrinkage phenomenon occurs in ceramic bodies during water removal in drying and sintering process which sometimes can result in physical defects in ceramic products such as crack and warping. Percentage of shrinkage and percentage of expansion for sintered double layers pellet between natural drying and microwave drying is shown in Fig. 2. Fig. 2 (a) shows that the double layers $\text{Al}_2\text{O}_3/\text{Al}_2\text{O}_3$ undergoes shrinkage while the double layers $\text{Al}_2\text{O}_3/\text{SiO}_2$ undergoes expansion after the sintering process. This graph shows that the microwave drying gives the lowest value of drying shrinkage for two similar materials, $\text{Al}_2\text{O}_3/\text{Al}_2\text{O}_3$. Typically, the drying process in microwave is more uniform due to the penetration depth [15, 20] of the microwave radiation and allows the shrinkage take place from the centre the surface of the material. As shown in Fig. 2 (a), double layers $\text{Al}_2\text{O}_3/\text{Al}_2\text{O}_3$ structure has a stable percentage for both drying method meanwhile the

percentage of expansion for double layer $\text{Al}_2\text{O}_3/\text{SiO}_2$ significantly different expansion between both of the techniques. The $\text{Al}_2\text{O}_3/\text{SiO}_2$ structure undergoes expansion due to the sintering temperature. This double layered structures were sintered at 1550°C (based on sintering temperature of Al_2O_3) and this causes the molten silica flow into the alumina structure. This is due to the fact that silica has slightly lower sintering temperature. In this case also the microwave drying gives a higher percentage of expansion compare to the natural drying technique due to the fact that this drying method also allows the particles stacking closed together which provide a good path for the diffusion process in the later stage of sintering process.



(a)



(b)

Fig. 2 (a) Percentage of shrinkage for double layers $\text{Al}_2\text{O}_3/\text{Al}_2\text{O}_3$ and (b) percentage expansion for double layers $\text{Al}_2\text{O}_3/\text{SiO}_2$

3.2 Porosity and Density

Table 1 displays the percentage of apparent porosity and bulk density for sintered double layers $\text{Al}_2\text{O}_3/\text{Al}_2\text{O}_3$ and $\text{Al}_2\text{O}_3/\text{SiO}_2$ structure. Table 1 shows that microwave drying produced higher porosity for sintered double layers $\text{Al}_2\text{O}_3/\text{Al}_2\text{O}_3$ structure compared to natural drying. Basically, shrinkage has an influence to the porosity

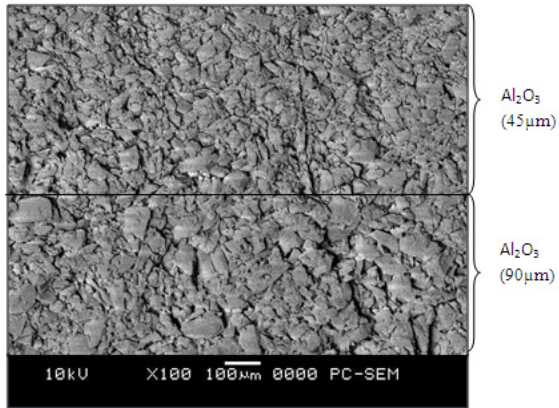
where an increase in shrinkage results in the reduction of porosity in the sintered ceramic bodies. As expected, the different double layers structure give different porosity and density. It showed that the natural drying produced the higher porosity and lower density for double layers $\text{Al}_2\text{O}_3/\text{SiO}_2$. Density and porosity are interrelated properties in ceramic materials. Theoretically, ceramic bodies with higher porosity will have a low density. The decrease of the bulk density is due to the increase of the porosity in sintered ceramic bodies. The finding reveals that shrinkage phenomenon is strongly related to the porosity and density.

Table 1 Porosity and density of double layers $\text{Al}_2\text{O}_3/\text{Al}_2\text{O}_3$ and $\text{Al}_2\text{O}_3/\text{SiO}_2$

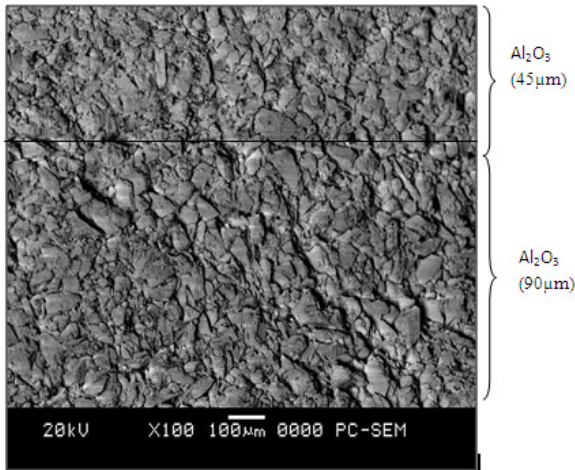
Property	$\text{Al}_2\text{O}_3/\text{Al}_2\text{O}_3$		$\text{Al}_2\text{O}_3/\text{SiO}_2$	
	Porosity (%)	Density (g/cm^3)	Porosity (%)	Density (g/cm^3)
Drying				
Microwave	33.47	3.60	26.55	2.85
Natural	31.85	3.69	27.50	2.79

3.3 Microstructural Analysis

SEM (Fig. 3 and 4) micrographs with EDS analysis (Fig.5) present the microstructure of sintered double layers structure $\text{Al}_2\text{O}_3/\text{Al}_2\text{O}_3$ and $\text{Al}_2\text{O}_3/\text{SiO}_2$. In natural drying, the particles are less agglomerated to each other and obviously create two separated layers for both cases while microwave offered uniform drying which caused the particle stacked closely together. This is clearly shown in Fig. 3(b) and 4(b) where the layer obviously separated for both double layers ($\text{Al}_2\text{O}_3/\text{Al}_2\text{O}_3$ and $\text{Al}_2\text{O}_3/\text{SiO}_2$) with the natural drying due to the uncontrolled drying process. Theoretically, the ceramic materials need a slow and controlled drying stage to avoid the cracking mechanism in ceramic green bodies. The structure obtained in Fig. 3 (a) and 4(a) clearly shown that both double layers structure have good linkage between layers with no crack and deformation in the sample. In natural drying, larger grains were obtained in the sintered double layered ceramic structure and this is opposite from the microwave drying where the application of microwave has successfully produced smaller grain in the sintered double layers structure. These result indicated that the microwave absorption characteristics vary depending on the microstructure where the finer and better microstructure can be produced by the microwave processing.

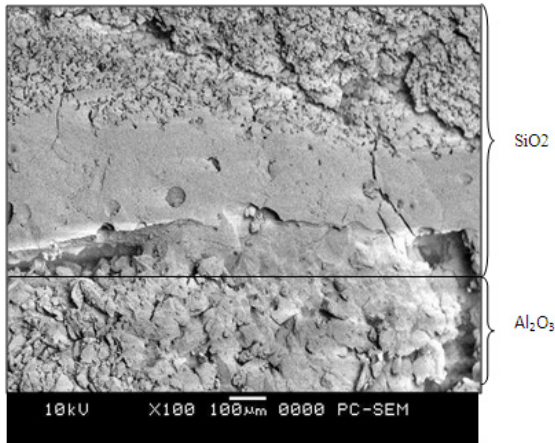


(a)

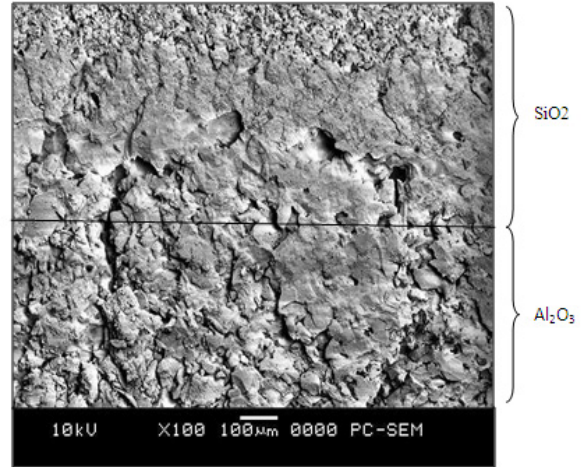


(b)

Fig. 3 Cross sectional images of sintered double layers ceramic structure ($\text{Al}_2\text{O}_3 / \text{Al}_2\text{O}_3$) that dried using different technique; (a) microwave, (b) natural

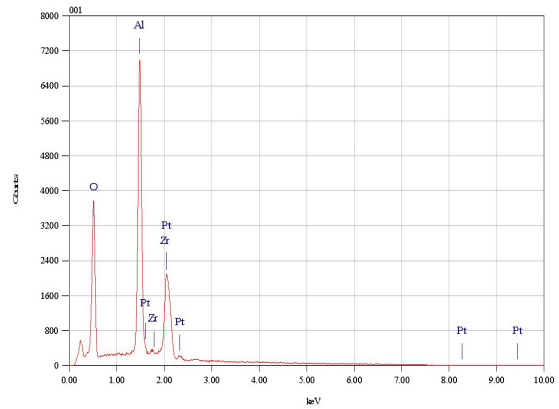
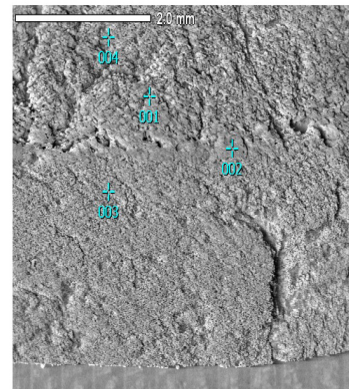


(a)



(b)

Fig. 4 Cross sectional images of sintered double layers ceramic ($\text{Al}_2\text{O}_3 / \text{SiO}_2$) that dried using different technique; (a) microwave (b) natural.



(a)

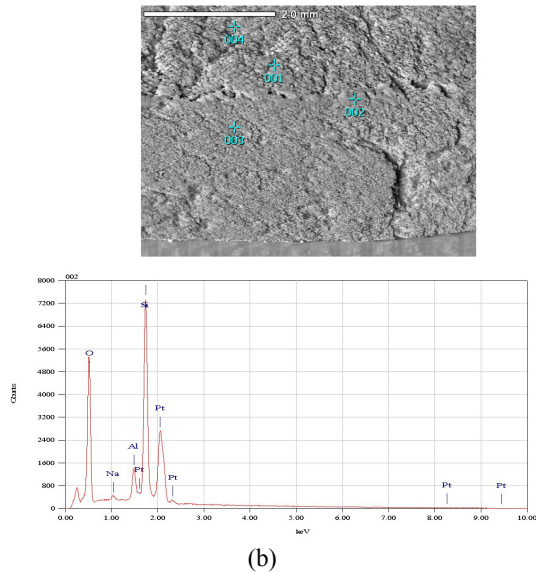


Fig. 5 EDS mapping for double layers ceramic Al₂O₃ / SiO₂

4. Conclusion

The application of microwave technique in drying double layers ceramic structure has potential to improve the properties of the structure. This is due to the several advantages of microwave drying such as shorter drying time which is good for thick samples especially for double layers structure and more uniform over the cross section part than other natural drying method. This is due to the penetration depth of the microwave radiation that can cause the water evaporated more uniform either at surface or centre. Meanwhile the uniform drying can lead to a homogenous temperature distribution and causes the shape of the sample maintains in the most of the structure. The finding of this study revealed that the microwave drying improves the properties of Al₂O₃/Al₂O₃ and Al₂O₃/SiO₂ double layered structures.

REFERENCES

[1] Lee H.J., Hiroyuki, S., and Kenji, H., (2005). Gas Permeation Properties in a Composite Mesoporous Alumina Ceramic Membrane, *Korean J. Chem. Eng.*, 22(5), p. 721-728.

[2] Anderson, B., and Riggard, E. The Effect of Ceramic Microstructure on the Properties and Durability of Multilayer Actuators, *SENSOR, Noliac. dk*. Retrieved 10th January 2011 from http://www.noliac.com/files/billeder/pdf/the_effect_of_ceramic_microstructure_on_the_properties_and_durability_of_multilayer_actuators.pdf.

[3] Janghorban K., (2009). Multilayer Diffusion Bonding of Polycrystalline Al₂O₃ to Alloys

Containing Sr, Mg and Si, *Journal of Science and Technology*,33(B),p.241-251.

[4] Somiya, S., Aldinger, F., Claussen, N., Spriggs, R. M., Uchino, K., Koumoto, K., and Kaneno, M.,(2003) *Handbook of Advanced Ceramics. Vol: 1.* Oxford: Elsevier Academic Press.

[5] Mujumdar, A.S., (2007). *Handbook of Industrial Drying, Third Edition Training*, CRC/Taylor & Francis.

[6] Gavrila, C., Andrian, G., and Gruia, I., (2008), *Heat and Mass Transfer in Convective Drying Processes*, COMSOL Conference 2008 Hannover.

[7] Kowalski, S. J., Grzegors, M., and Jacek, B., (2009), *Heat and Mass Transfer during Microwave-Convective Drying*, Wiley Interscience, *AIChE Journal*,56(1), p.24-35.

[8] Khodaparast,H., Akhbar, and Ghanadzadehan, H., (2005). *Experimental Survey on Microwave Drying Porous Media*, *Iran. J. Chem.Eng.*, 24(2).

[9] Rahman, H. A., Shariff, M., and Rahmat, N. A., (2009). *A Comparative Study of Green Alumina Foams Prepared by Microwave and Conventional Drying*, 4th International Conference on Recent Advances in Materials, Minerals & Environment and 2nd Symposium on Materials & Processing 2009,1-3 June 2009, Penang, Malaysia.

[10] Thostenson, E.T., and Chou, T.W., (1999). *Microwave Processing: Fundamental and Applications*, *Composites:Applied Science and Manufacturing*, 30, p.1055-1071.

[11] Lefeuvre S., (1981). *Microwave Drying of Porous Materials*, *Physics Technology*, 12.

[12] Skansi, D., and Tomas, S., (1995). *Microwave Drying Kinetics of a Clay Plate*, *Ceramic International* 21, p.207-211.

[13] Ratanadecho P., Aoki, K., and Akahori, M., (2002). *Influence of Irradiation Time, Particles Sizes and Initial Moisture Content During of Multilayered Capillary Porous Materials*, *Journal of Heat Transfer*, 124(1), P.151.

[14] Meredith, R., (1998). *Engineers' of Industrial Microwave Heating IEE Power Series 25*, The Institution of Electrical Engineering, London, United Kingdom.

[15] Rahman, H. A., Rahmat, W., and Shariff, M. (2009). *Microwave Drying Effect on the Properties of Alumina Zeolite Foam*, 2nd Annual International Conference on Green Technology and Engineering (ICGTE 2009), Lampung, Indonesia.

[16] Zhipeng, X., Jinlong, Y., Xiangdong, H., and Yong, H., (1999). *Microwave Processing and Properties of Ceramic with Different Dielectric Loss*, *Journal of the European Ceramic Society* 19, p.381-387.

[17] Oloyede, A., and Groombridge, P., (2000). *The influence of microwave heating on the mechanical properties of wood*, *Journal of Materials Processing Technology*, p.67-73.

[18] Hiroyuki, M., Mikinori, H., Kita, H., and Izutsu, Y., (2011). *Joining of Alumina with a Porous Alumina Interlayer*, *Journal of Ceramic International*, National

- Institute of Advanced Science and Technology (AIST), p.7.
- [19] Kang, L., (2007). Ceramic membranes for separation and reaction, John Wiley and Sons, Ltd
- [20] Brosnan, D. A., and Robinson, G. C., (2003). Introduction to drying of ceramics with laboratory Exercises. The American Ceramic Society