CAPILLARY VELOCITY OF NATURAL ZEOLITE POROUS CERAMIC IN DIFFERENT SINTERING TEMPERATURES

Sri Mulyo Bondan Respati^{1,2}, Rudy Soenoko², Yudy Surya Irawan² Wahyono Suprapto², Wahid Budi Saputra¹, and Helmy Purwanto¹ ¹Wahid Hasyim University, Mechanical Engineering Department Semarang, Indonesia

> ²Brawijaya University, Mechanical Engineering Department Malang, Indonesia

DOI: 10.17973/MMSJ.2017_06_2016104

e-mail: bondan@unwahas.ac.id

Lately, the membrane of porous ceramics begin to increase its utility since it contains good and supportive chemical substances, besides it is also resistant to heat and has stable force. However, its price is considered quite expensive because this type of ceramics is made from materials and is produced through complicated process. This study aims at utilizing the use of natural zeolite materials as the material to use for membranes which offers low price. This material can be used as a membrane because of its unique physical and chemical characteristics and it contains liquid adsorbent. In this study, the natural zeolite was obtained from Malang, Indonesia. Porous ceramic sintering is a process of ceramics making by baking the ceramic at the temperature of around 700-900 °C. In this study, the porosity and capillary velocity cylindrical porous ceramic were investigated. Density and porosity of the material were measured using the Archimedes method (ASTM C20). The capillary absorption coefficient was calculated using the standard MSZ EN 1925: 2000. The result of this study shows that the higher the sintering temperature, the greater the density and the smaller the open porosity produced in the process which increases the capillary velocity. This phenomena occured due to the greater driving force which makes the pores of porous ceramics made of natural zeolite and the capillary velocity is increased.

KEYWORDS

ceramic, sintering, natural zeolite, porous, capillary

1. INTRODUCTION

Lately, ceramic membranes have been widely used for its ability to drain small droplets of fluid. To drain tiny water droplets in quite small amount of certain fluid, it is necessary to use small hollow ceramics. Small cavities are expected to be able to hold the metal content that has larger sizes in the water. Mostly, in the market of ceramic, the membranes made of alumina and zirconia were considered expensive since the manufacturing process or the materials required long and expensive process. Recently, zeolite materials have been introduced in ceramics industry for its ability to reduce heavy metal substances, Susanawati, et al. [2011]. For that there needs to research the manufacture of ceramics made from natural zeolite is used as a membrane. Because when the use of zeolite for water filtration more use of large grains, this grain are that can move to produce a large cavity as well.

Research on ceramic membrane of zeolite materials have been done by previous researchers such as Histrov, et al. [2012] which in his study, he used natural zeolite material from Bulgaria. The result of his study showed that ceramics can be sintered at temperature of between 800–1000 °C. At the temperature of between 800–900 °C, porosity still occurred while at the temperature of 1000 °C, the porosity no longer occured. San et al. [2003] San and Ozqur [2011] examined the use natural zeolite with a mixture of quartz at a temperature of 950 and 1 000 °C. The result of their study showed that there were pores in the ceramic and they explained that the zeolite began to melt at the temperature 800 °C. Respati, et al. [2016] had measured ceramic porosity zeolite using Archimedes' principle and two-dimensional porosity on sintering at the temperature of 800, 850, and 900°C. The compacting pressure on the green part was set at the pressure of 25 MPa. The result of this study explained that there was porosity of around 28.13-47.61 %. Respati, et al. [2014] have performed sintering alumina ceramic with mixed zeolites. The percentage of the amount of zeolite at 10–40 % affected shrinkage of alumina. When the zeolite was added with alumina, it enlarged the ceramic shrinkage. In those studies, researchers still found the existence of porous ceramics. The porous ceramics, which are used as the membrane should be able to absorb water. Furthermore, water absorption can be determined by the capillary level of the material.

The capillary level is very important in a porous ceramic since it indicates that the porous ceramic can be irrigated [Juhasz et al. 2014]. Thus, under certain conditions, it can be used as a membrane to filter the water through the application of the zeolite ceramic.

This study aimed at exploring the characteristics of ceramics made from natural zeolite. The natural zeolite material was obtained from miners zeolite Malang, Indonesia. The natural zeolite ceramic has the characteristic of being sought to shrinkage, density, porosity and velocity capillary.

2. EXPERIMENTAL MATERIALS

Materials used in this study were the natural zeolite from Malang, Indonesia with the composition of chemical analysis test of: 66.9 % SiO₂, 11.43 % Al₂O₃, 4.59 % Fe₂O₃, 0.18 % TiO, 2.40 % CaO, 1.44 % MgO, 1.95 % K₂O, 1.29 % Na₂O.

3. EXPERIMENTAL METHODS

Natural zeolite powder was screened with a 120 mesh sieve. Zeolite powders under 120 mesh was then molded into a green body specimens. The cylinder specimen shape with a diameter of 10 mm and a height of 20 mm is shown in Fig. 1. Compacting pressure on green body pressure was set at 40 MPa. The heating process was done using an electric furnace which temperatures were set at 700, 750, 800, 850 and 900 °C, with heating rate of 5 °C per minute and a holding time of 6 hours. The sintering temperatures of 700–900 °C were used considering the reason that when the sintering temperature limit of the porous zeolite ceramic goes more than 1 000 °C, it will result to solid ceramic that have no porosity and cannot be irrigated. Specimens were measured using Archimedhes' method (ASTM C20) [Berger 2010]. Capillary absorption coefficient calculation was done using the standard MSZ EN 1925: 2000 [Juhasz et al. 2014].



Figure 1. Dimension green part specimen

Shrinkage was calculated through the ceramic volume data and the volume of green parts data. The equation used to measure was the equation 1.

$$s_V = \frac{v_g - v_c}{v_g}$$
(1)

Where V_g is the volume of the green part and V_c is the volume of ceramics. The shrinkage of weight measurement employed the equation 2.

$$e_W = \frac{w_g - w_c}{w_g}$$
(2)

Where W_{g} is the weight of green parts and W_{c} is the weight of the ceramic. The bulk density was calculated using equation 3.

$$\rho_{bulk} = \frac{W_{in air}}{V_{total}}$$
(3)

Where $W_{in air}$ is the ceramic weight in the air and V_{total} is ceramic bulk volume including porosity. As for the apparent density was calculated using Equation 4.

$$\rho_{apparent} = \rho_{water} \times \frac{W_{in air}}{W_{in air} - W_{in water}}$$
⁽⁴⁾

Where $W_{in water}$ is the ceramic weight soaked in the water and ρ_{water} is the density of water. Open porosity calculated using equation 5.

$$P_{open \ porosity} = 1 - \frac{v_{app \ arent}}{v_{total}} \times 100\%$$
 (5)

Where $V_{\mbox{\tiny apparent}}$ is the real volume and how to find $V_{\mbox{\tiny apparent}}$ was using the equation 6.

$$V_{apparent} = \frac{W_{in air} - W_{in water}}{\rho_{water}}$$
(6)

The procedure of measuring the capillary velocity was done by dipping the tip of the specimen on the surface of the water. The time used to do the process of creeping water until the water soaks specimen was measured recorded. The capillary velocity was calculated by dividing the high off the water soaks specimen by the time required to do the process.

4. RESULTS

The average bulk volume of the green body specimens at a sintering temperature of 700, 750, 800, 850 and 900 °C respectively were found at 2.53, 2.48, 2.48, 2.40, and 2.49 cm³ while the average bulk volume of ceramics were 2.45, 2.37, 2.35 , 2.18, and 1.91 cm³. A volume shrinkage in a row were 3.37, 4.60, 5.01, 9.30, and 23.41%. While the shrinkage of the weight were 5.67, 5.93, 6.42, 9.70, and 13.82%. The average bulk density zeolite ceramic in a row were 1.22, 1.25, 1.25, 1.43, and 1.43 g/cm³. Value shrinkage is presented in the graph as shown in Fig. 2. Shrinkage increases at was the sintering temperature increases and begins to rise sharply at the temperatures of around 800–900 °C.



Figure 2. The correlation between sintering temperature and shrinkage

This shrinkage occurs because the sintering temperature increased, making the driving force to shrink pores becomes greater. This shrinkage occurs because the increased the sintering temperature creates a driving force to shrink the pores to become larger [Li and Pan 2013]. Shrinking pores were then followed by the process of letting out the air from grain which makes the specimen shrink. This shrinkage happens due to the increasing density of the ceramics. This was evidenced by the density measurement.

By using the Archimedhes method, apparent density at sintering temperatures of 700, 750, 800, 850, and 900 °C were obtained and resulted to the average of apparent density at 2.21, 2.22, 2.23, 2.24, and 2.38 g/cm³. Graph bulk density and the apparent density are illustrated in Fig. 3. This graph shows that the higher the sintering temperature the higher the density which increased sharply at a temperature of 900 °C.



Figure 3. Relationhip between the sintering temperature and density

The results of density measurements indicate that when the increasing natural zeolite ceramic meets with the increasing density, it will produce stronger ceramics. In relation to the stronger bond, the decreased porosity ceramic also results to decreased strength. To determine the open porosity, the researchers employed the Archimedes principle to measure it. Open porosity refers to the porosity which is associated with the surface. It is believed that the decrease on the open porosity will affect the pores to be decreased into smaller sizes.

Open porosity was calculated for each successive temperature of 44.8, 44.2, 44.1, 43.9, and 40.1 %. Fig. 4. describes the increase on sintering temperature causing smaller porosity which falls sharply at a temperature of 900 $^{\circ}$ C.



Figure 4. Relation of sintering temperature and porosity

The decreased open porosity indicates that the holes in the ceramic are getting the fewer in amount or smaller in size. These results go in line with the result of the research done by Histrov et al. [2012] and Respati et al. [2016]. In this study, we tried to add one more testing variable which was the capillary velocity which had not yet connected with the porosity in previous research. If the size of the smaller pores will then make the absorption of water more quickly intervening even though the water content less. The speed of water absorption can be measured through the capillary velocity.

The results of the measurement on capillary velocity were obtained successively 2.30, 2.32, 2.38, 3.37, and 5.31 mm/min as shown by the chart in Figure 5. Meanwhile, he capillary velocity goes faster with increasing sintering temperature and the temperature begins to rise sharply at the temperature of 800–900 °C.

With the increasing of the velocity capillary it was expected that the holes were very small pores that can filter out dirt water that were larger than the pores hole. Finally, the effect of temperature sintering were the sintering temperature increases, then make the size smaller pores so velocity capillary were also increased.



Figure 5. Relationship of the sintering temperature and capillary velocity

Fig. 6 describes the porosity which influences the capillary velocity. The greater the porosity the larger the pores, making the absorption of the water becomes slow. Therefore, to make a rapid fluid flow which is better in filtering heavy iron impurities, it is required to create a membrane with small holes in larger area. The material weight will become heavier due to the volume shrinkage which are greater than the shrinkage weight.

5. CONCLUSION

This study showed that the effect of increasing sintering temperature makes the natural zeolite ceramics density and porosity decreased, yet it slit pores smaller which increases the absorption force. The result of this study suggests that the temperature sintering of natural zeolite ceramic should be set at around 700–900 °C to perform better in water absorption. The high increase on the sintering temperature will make the size of the pores becomes smaller, making it possible for the water to be well absorbed since the particles of the water are smaller than the pores.

References

[Berger 2010] Berger, B. The Importance and Testing of Density/ Porosity/ Permeability/ Pore Size for Refractories, Proceeding of The Southern African Institute of Mining and Metalurrgy Refractories Conference, pp 101-116

[Hristov et al. 2012] Hristov, P., Yoleva, A., Djambazov, St., Chukovska, I., Dimitrov, D. Preparation And Characterization Of Porous Ceramic Membranes For Micro-Filtration From Natural Zeolite, *J. the University of Chemical Technology and Metallurgy*, 2012, Vol. 47, No. 4. pp 476-480, ISSN 1314-7471

[Juhasz et al. 2014] Juhasz, P., Kopecsko, K., Suhajda, A. Analysis of capillary absorption properties of porous limestone material and its relation to the migration depth of bacteria in the absorbed biomineralizing compound, *Per. Pol Civil Eng*, 2014. Vol. 58, No. 2, pp 113-120, ISSN 0553-6626



Figure 6. Relationship between porosity and capillary velocity

[Li and Pan 2013] Li F., and Pan, J. Modelling "Nano-Effects" in Sintering, in: R.H.R. Castro and K. van Benthem (eds.), Sintering, *Engineering Materials*, Vol. 35, Springer-Verlag Berlin Heidelberg, 2013, ISBN 978-3-642-31008-9

[Respati, et al. 2014] Respati, S. M. B., Soenoko, R., Irawan, Y. S., Suprapto, W. Effect of Weight Percentage Natural Zeolite on Alumina Natural Zeolite Ceramic Matrix Composites Shrinkage (in Indonesian), Proceeding of National Seminar on Science and Technology 5, Semarang, 25 Juni 2014, Semarang: Faculty of Engineering, Wahid Hasyim University, pp 90-94, ISBN 978-602-99334-3-7

[Respati et al. 2016] Respati, S. M. B., Soenoko, R., Irawan, Y. S., Suprapto, W. Effect of Low Temperature Sintering on the Porosity and Microstructure of Porous Zeolite Ceramic, *Applied Mechanics and Materials*, June 2016, Vol. 836, pp 219-223, ISSN: 1662-7482

[San et al. 2003] San, O., Abali, S., and Hosten, C. Fabrication of Microporous Ceramics from Ceramic Powders of Quartz-Natural Zeolite Mixtures, Ceramic International, 2003, Vol. 29, pp 927-931, ISSN 0272-8842 [San and Ozgur 2011] San, O., and Ozgur, C. Sintering Effect on the Microstructure of Glassy Porous Ceramics, 6th IATS'11, 16–18 May, Elang Turkey, 132-135,

[Susanawati et al. 2011] Susanawati, L. D., Suharto, B., and Kustamar Decrease content of Heavy Metals in Water Leachate by Zeolite Media Method Using Batch and Continuous Method (in Indonesian), J. Agrointek, October 2011, Vol. 5, No. 2, pp 126-132, ISSN 1410-4121

Contacts

Sri Mulyo Bondan Respati, ST. MT Wahid Hasyim University Faculty of engineering JI. Menoreh Tengah X/22, Sampangan 50236 Semarang, Indonesia tel: +62 24 8506680 e-mail: bondan@unwahas.ac.id