Fabrication and characterization of fine ceramic based on alumina, bentonite, and glass bead

P Sebayang¹, Nuraida², S Simbolon¹, C Kurniawan¹, M Yunus¹, E A Setiadi¹ and Z Sitorus²

¹Research Center for Physics, Indonesian Institute of Sciences, Kawasan PUSPIPTEK, Tangerang Selatan, Indonesia
²Physics Department, Faculty of Mathematics and Natural Science, University of North Sumatera, Medan, Indonesia

E-mail: pard003@lipi.go.id

Abstract. Fabrication of fine ceramics based on alumina, bentonite and glass bead has been carried out by powder metallurgy. The preparation of powder has been performed using High Energy Milling (HEM) with wet milling process and using toluene as medium for 2 hours. The powder milling result was dried in oven at 100 °C for 24 hours. After that, the powder was compacted into pellet by using hydraulic press with 80 kgf/cm² pressure at room temperature. Then, the pellet was sintered at 900 °C for 4 hours. Materials characterization such as physical properties (true density, bulk density, porosity, and water absorption), average particle diameter, hardness, microstructure and phase were measured by Archimedes method, Particle Size Analyzer (PSA), Hardness Vickers (HV), Scanning Electron Microscope (SEM-EDX) and X-Ray Diffraction (XRD). From the result, the optimum condition is sample D (with addition of 30 wt.% γ-Al₂O₃) with sintering temperature of 900 °C for 4 hours. At this condition, these properties were measured: average particle diameter of 4.27 µm, true density of 2.32 g/cm³, porosity of 5.57%, water absorption of 2.46%, bulk density of 2.39 g/cm³, and hardness of 632 HV. The fine ceramic has four phases with albite (Al₂NaO₆Si₃) and quartz (SiO₂) as dominant phases and corundum (Al₂O₃) and nepheline (AlNaO₄Si) as minor phases.

1. Introduction
Ceramics can be classified into traditional and fine ceramics. Traditional ceramics are generally made from naturally occurring raw materials such as clay, mica, quartz, porcelain, and pottery. Most of the traditional ceramics are based on porous silicates whose microstructures are not homogeneous and consist of many phases. Meanwhile, the fine ceramics are mostly developed through synthesis process or powder metallurgy. The fine ceramics can be produced predominantly from alumina, titanium, zirconia, magnesia, and other oxides [1, 2]. Alumina (Al₂O₃) is an oxide or fine ceramics that can be applied in various fields such as electronic and mechanical component [3].

The Al₂O₃ ceramics has melting point of about 2000 °C, density of 3.96 g/cm³, and hardness of 14 kg/mm². The Al₂O₃ ceramics is electrically insulating, strong, hard, and resistant to high temperatures [4]. However, the growth of alumina ceramic grains becomes abnormal at high temperatures. This causes inhomogeneous microstructures of formed grains which can degrade the quality of ceramics [5]. One way to lower the sintering temperature is to reduce grain sizes into nanoparticle or
to incorporate certain additives which have lower melting point than alumina, such as MgO and SiO$_2$ [6]. An addition of silica (SiO$_2$) materials to alumina ceramics can form a liquid phase and enable to repair densification of the materials. Excessive addition of SiO$_2$ will impact to the decreasing mechanical strength on alumina ceramics [7]. Besides an addition of silica, bentonite can also be used as a mixture to enhance the flexibility of ceramic materials.

The bentonite is a type of clay with 85% montmorillonite minerals and 15% others such as cristobalite, feldspar, calcite, gypsum, kaolinite, plagioclase, and illite [8]. The chemical formula of bentonite is (Na, Ca) O.Al$_2$O$_3$.4SiO$_2$.H$_2$O [9]. The special properties of bentonite are the ability to form thixotrophic gel with water, high capacity of exchange cation, and to absorb water in large quantity until it increases its volume around 12-15 times. The ability of montmorillonite to absorb water will decrease after heating at critical temperature at 105 – 390 °C [8].

The manufacture of fine ceramics based on bentonite, SiO$_2$, and γ-Al$_2$O$_3$ powders is widely used as ceramic filter and furnace equipment material. Besides, glass bead is also used as additive of ceramics. The addition of glass bead can improve the thermal and mechanical properties of ceramics [10].

In this research, we studied the fabrication of fine ceramics based on bentonite, glass bead, and γ-Al$_2$O$_3$ powders. The ceramic was prepared by metallurgy method with wet milling process. It was compacted into pellet by die pressing method and sintered at 900 °C for 4 hours [11]. The manufacture of fine ceramics based on bentonite, glass bead, and alumina is expected to improve the physical and mechanical properties of this material.

### 2. Experimental method

Bentonite, glass bead, and γ-Al$_2$O$_3$ powders were used as raw materials and the compositions are shown in table 1.

| Sample code | Composition (wt.%) |
|-------------|--------------------|
|             | Alumina  | Bentonite | Glass bead |
| A           | 0        | 50        | 50         |
| B           | 10       | 45        | 45         |
| C           | 20       | 30        | 30         |
| D           | 30       | 35        | 35         |

The fabrication of fine ceramics was carried out by powder metallurgy method. The raw materials were mixed and milled by using High Energy Milling (HEM). This process was done by wet milling for 2 hours and using toluene as liquid medium. Comparison of ball mill to powder was fixed at 10:1. After the milling process, the powder was dried in the oven at 100 °C for 24 hours. Furthermore, the powder was compacted into pellet with pressure of 80 kgf/cm$^2$ at room temperature. The pellet was then sintered at 900 °C temperature for 4 hours.

Characterizations of the materials were performed including true density, bulk density, porosity, and water absorption using Archimedes method [12, 13]. The true density measurement of the powder mixture was carried out using Pycnometer. While the average particle diameter was measured using Particle Size Analyzer (PSA) Cilas-1190, hardness (HV) was performed using Micro Hardness Tester (LECO LM-100AT), morphological using Hitachi SU-3500 Scanning Electron Microscopy (SEM), and phase was analyzed by using X-Ray Diffraction (XRD) Rigaku Smart Lab type, λ = 1.5418 Å.

### 3. Results and discussion

Diameter distribution results of the powder mixture are shown in figure 1. The diameter powders of the samples are 5.86, 5.17, 5.13, and 4.27 µm with various alumina additions of 0, 10, 20, and 30 wt.%, respectively. It shows that the variation of alumina addition (Al$_2$O$_3$) does not give any effect to
the particle size of resulted sample. The same particle size of sample shows that the milling time of the fine ceramic fabrication is optimum.

![Graphs showing particle size distribution](image)

**Figure 1.** Particle size distribution after wet milling with various addition of alumina (Al$_2$O$_3$): (a) 0, (b) 10, (c) 20, and (d) 30 wt.%.

Figure 2a shows the percentage of alumina addition (Al$_2$O$_3$) versus bulk and true density, while figure 2b shows porosity and water absorption. The bulk density value is around 2.16 - 2.39 g/cm$^3$, while true density value is around 2.28 - 2.32 g/cm$^3$. The porosity value is obtained in range of 5.57 - 9.82% and water absorption is around 2.46 - 5.01%. The effect of alumina addition (Al$_2$O$_3$) tends to increase true and bulk density values. Alumina has higher true density than bentonite (true density of alumina = 3960 kg/m$^3$, true density of glass bead = 120 kg/m$^3$, and true density of bentonite = 25 – 40 kg/m$^3$), so the variation of alumina addition contributes to the change of density. In the other hand, from figure 2b it can be seen that the addition of alumina decreases porosity and water absorption. The porosity is inversely proportional with the density value. As the density value increases, the porosity of the sample decreases.

SEM-EDX results are shown in figure 3. Figures 3a and 3b are for Sample A, and 3c and 3d for Sample D. The aluminum element Al increases with the addition of $\gamma$-Al$_2$O$_3$. In Sample A, it has flake-like morphology with porous structure. The interlayer spaces of bentonite, which was thermally activated, can be collapsed and result in more tightly bound structure with reduction in porous structure [14]. However, in this case the addition of glass beads decreases the porosity of sample, and the surface becomes rather flat caused by melting of glass bead. The dominant elements composition of Sample A (without addition of $\gamma$-Al$_2$O$_3$) are O = 79.4 at.%, Si = 13.18 at.%, Na = 4.15 at.%, and Al...
Figure 2. The effect of percentage of alumina addition (Al₂O₃) to (a) average particle diameter and true density and (b) porosity and water absorption.

Figure 3. SEM-EDX results of (a) element composition and (b) morphology from Sample A. (c) Element composition and (d) morphology from Sample D.

The result of fine ceramics microstructure analysis using X-Ray Diffraction (XRD) is shown in figure 4. The performed analysis was limited before and after the addition of 30 wt.% Al₂O₃ with sintering temperature of 900 °C for 4 hours. Figure 4 shows that with or without addition of 30 wt.% Al₂O₃ the ceramic has four phases, which are albite (Al₂NaO₅Si₃), corundum (Al₂O₃), nepheline
(AlNaO₄Si), and quartz (SiO₂). This result is consistent with EDX analysis. Before sintering temperature, no nepheline phase was observed. As the alumina was being added, the new phase of nepheline was formed. We suggest that the excess of additive alumina and the existence of silica in bentonite and glass bead tend to form nepheline than albite.

Figure 4. X-ray diffraction pattern of Samples A and D.

The Al₂NaO₈Si₃ phase has a triclinic crystal structure with parameters a = 8.259 Å, b = 12.975 Å, and c = 7151 Å. Al₂O₃ phase has a trigonal crystal structure with lattice parameters a = 4.766 Å and c = 12.980 Å. AlNaO₄Si phase, it has a hexagonal crystal structure with lattice parameters a = 9.995 Å, c = 24.797 Å. SiO₂ phase has a hexagonal structure with lattice parameters a = 4.918 Å, c = 5.470 Å. The dominant phases of the fine ceramic are albite (Al₂NaO₈Si₃) and SiO₂, while the corundum (Al₂O₃) and nepheline (AlNaO₄Si) are minor phase.

Figure 5 shows the effect of the percentage of alumina addition (Al₂O₃) to hardness (HV) on the manufacture of fine ceramics. The obtained hardness value increases from 495 to 632 HV. This is due to alumina, which has relatively higher hardness [4]. Therefore, an addition of Al₂O₃ results in increasing of fine ceramics densification. The reason is that during the sintering process heat energy activated diffusion process between the granules, therefore the densification process occurs followed by volume reduction but not by mass change. The optimum condition is achieved by addition of 30 wt.% Al₂O₃ and sintering temperature of 900 °C for 4 hours [11], resulting in average particles diameter of 4.23 μm, true density of 2.32 g/cm³, porosity of 5.57%, water absorption of 2.46%, bulk density of 2.39 g/cm³, and hardness of 632 HV. The optimum sample is obtained at the higher hardness condition.

Figure 5. The effect of alumina (Al₂O₃) addition to bulk density and hardness (HV).
4. Conclusions
Fine ceramics based on alumina (γ-Al2O3), bentonite, and glass bead have been successfully fabricated using HEM with wet milling process. The resulting powder from 2 hours milling has average diameter of 4.23 μm and true density of 2.32 g/cm³. The optimum condition is obtained in Sample D with the addition of 30 wt.% Al2O3 and sintering temperature of 900 °C for 4 hours because it has the higher hardness. The properties of Sample D are: porosity of 5.57%, water absorption of 2.46%, bulk density of 2.39 g/cm³, and hardness of 632 HV. Fine ceramics produced from alumina material (γ-Al2O3), bentonite and glass bead have dominant phase albite (Al2NaO8Si3) and quartz (SiO2), and minor phases corundum (Al2O3) and nepheline (AlNaO3Si).

Acknowledgments
The authors acknowledge to P2F - LIPI 2017 for fund and characterization facility.

References
[1] Rasin F A and Hamad E A 2012 British Journal of Science 7 56
[2] Heimann R B 2010 Classic and Advanced Ceramics: From Fundamentals to Applications (Weinheim: WILEY-VCH Verlag GmbH & Co. KGaA)
[3] Buchanan R C 1986 Ceramic Materials for Electronics (New York: Marcel Dekker, inc)
[4] Davis K 2010 School of Doctoral Studies (European Union) Journal 109
[5] Montanaro L, Tulliani J M Perrot C and Negro A 1997 J. Eur. Ceram. Soc. 17 1715
[6] Rhamdhani M A, Soepriyanto S, Ramelan A and Barliansya A 2005 Jurnal Teknologi Mineral 12 148
[7] Louet N, Reveron H and Fantozzi G 2008 J. Eur. Ceram. Soc. 28 205
[8] Adamis Z and Williams R B 2005 Bentonite, Kaolin, and Selected Clay Minerals Environmental Health Criteria vol 231 (Geneva: World Health Organization)
[9] Alemdar A, Öztekin N, Erım F B, Ece Ö I and Gungör N 2005 Bull. Mater. Sci. 28 287
[10] Huang L, Yuan Q, Jiang W, An L, Jiang S and Li R K Y 2004 J. Appl. Polym. Sci. 94 1885
[11] Chen Y F, Wang M C and Hon M H 2004 J. Eur. Ceram. Soc. 24 2389
[12] Kurniawan C, Nainggolan M M, Sebayang K, Ginting M and Sebayang P 2017 J. Phys.: Conf. Ser. 817 012057
[13] Setiadi E A, Kurniawan C, Sebayang P and Ginting M 2017 J. Phys.: Conf. Ser. 817 012054
[14] Toor M and Jin B 2012 Chem. Eng. J. 187 79