The characterization of ceramic alumina prepared by using additive glass beads

Suprapedi\textsuperscript{a}, Muljadi\textsuperscript{b}, Priyo Sardjono

Research Center for Physics-Indonesian Institute of Sciences, Kawasan Puspiptek Serpong Tangerang Selatan
a)mulj004@lipi.go.id
b)supr002@lipi.go.id

Abstract. The ceramic alumina has been made by using additive glass bead (5 and 10 \% wt.). There are two kinds of materials, such as: gamma Alumina and glass bead. Synthesis of alumina was done by ball milling for 24 hours, then the mixed powder was dried in drying oven at 100 oC for 6 hours. Furthermore, the dried powder was mixed by using 2 \% of PVA and continued with compacted to form a pellet with pressure of 50 MPA. The next step is sintering process with variation temperature of 1150, 1200, 1250, 1300 and 1400 oC and holding time for 2 hours. The characterization conducted are consist of test density, hardness, shrinkage, and microstructure. The results show that ceramic alumina with addition of 10 \% wt. glass bead has the higher value of density, hardness and shrinkage than addition of 5\% wt. glass bead. The highest characterization of ceramic alumina with addition 10 \% glass bead was achieved at sintering temperature of 1400 oC with density 3.68 g/cm\textsuperscript{3}, hardness vickers 780.40 Hv and shrinkage 15.23 \%. The XRD results show that it was founds a corundum (alpha Alumina) as dominant phase and mullite as minor phase.

1. Introduction
Alumina is one of the most important ceramic oxides and has a wide range of uses, including high-temperature applications (refractory materials), electrical insulator and microelectronics. Alumina is also important from the point of view that it has many meta stable polymorphs such as \(\kappa\), \(\gamma\), \(\delta\), \(\Theta\)-\(\text{Al}_2\text{O}_3\) and stable form \(\alpha\text{Al}_2\text{O}_3\) [1]. Among the several meta stable polymorphs of alumina (transition alumina phases), \(\gamma\)-alumina is important due to its application in industry as catalyst, adsorbents, coatings, and soft abrasives. A stable form of \(\alpha\text{Al}_2\text{O}_3\) are widely used in blast finishing and surface preparation due to their hardness, cost and longevity [1].

Alumina is the most widely used oxide ceramics because of its hardness, good corrosion resistance, high insulation and ease of processing [1,2]. Furthermore, its thermal expansion coefficient is nearer to those of metals than that of structural nitrogen ceramics, such as silicon nitride (\(\lambda\)\text{Iron} = 15 \times 10\textsuperscript{-6} \text{oC}^{-1}, \lambda\text{Si}_3\text{N}_4=3 \times 10\textsuperscript{-6} \text{oC}^{-1}) [3]. Therefore, if the mechanical properties of alumina are further improved, it is reasonable to expect that alumina could be widely used for engine part [3]. There are some steps for manufacturing of ceramic product, sintering process is one of the most important and crucial part in fabricating ceramic component. In fact, most of the ceramic component that fabricated via plastic forming and powder route for the variety applications must undergo this sintering steps [4]. Ceramic alumina has a very high melting point of about 2040\textdegree C, therefore to produce alumina ceramic product with maximum density requires sintering temperature near the
melting point, which is about 1700 - 1800°C [5,6]. There are two general approaches to enhance sintering process or to lower the sintering temperature. The first is to improve powder processing, that is, to use fine starting powders and to eliminate agglomerates in the green pre forms such as by chemical routes [7]. The second approach is to use sintering aids or additives [7]. Additives in solid solutions can enhance diffusion and hence sintering by increasing defect, while additives forming a liquid phase can facilitate particle rearrangement and solution / reprecipitation [8,9]. Some of additives of sintering were used commonly such as: SiO$_2$, Borax, MgO, TiO$_2$, Y$_2$O$_3$ and so on [10]. M. Sathiyakumar et all have reported that beneficial effect of small addition of MgO in alumina powder in achieving full density and even translucency by sintering and prevention of abnormal grain growth in the final stage of densification [11]. Glass beads are made of silica and other minerals melted at a elevated temperature to form a thick, viscous liquid. The liquid is molded into the desired shape and hardens as it cools. Glass beads are made in many sizes and shapes [12]. Commonly, beads are manufactured by winding molten glass around a long iron rod. Different shapes can be created while the bead is still semi soft. For instance, square and oval beads can be manufactured by pressing the bead while it is still hot against a flat surface. Shapes that are more intricate are manufactured with molds. Major oxides in most glasses comprise Na$_2$O, MgO, SiO$_2$, Al$_2$O$_3$, K$_2$O, and CaO, whereas other oxides are usually minor and trace compositions of the glass. Glass beads are generally glassy materials such as glass windows that have an amorphous structure with the main content of silica (SiO$_2$), glass beads containing alkali elements such as Ca, Na and K. Glass beads are formed in form a granule with a size of about 0.1 - 0.3 mm. Generally, the composition of glass bead consists of 18 ± 2% Na$_2$O, 67 ± 3% SiO$_2$ and 9 ± 2% CaO [12, 13]. In this study we used a glass bead as an alumina sintering additive. The purpose of this research is to find out the influence of addition of a glass bead additive composition to the sintering of alumina ceramic.

2. Experimental works
The starting material was a high-purity (>99.99%) γ-alumina powder with an average particle size of 10 µm and Glass bead as additive. The additive composition expressed in weight percent was 5 % and 10 %. Both of raw materials were milled by ball milling for 24 hours with using pure Al$_2$O$_3$ ball and water as milling media. Then the slurry was dried at 100°C and the dried powder was hand ground using mortar and pestle, then the fine powder was mixed with 2 % wt. of binder polyvinyl alcohol (PVA). Pellet samples with 12 mm in diameter were prepared by pressing under pressure 50 MPa using stainless steel die. The pellets were then dried for 24 hours under room temperature and sintering were conducted at variation temperature namely 1150, 1200, 1250, 1300 and 1400°C for 2 hours with holding time and heating rate of 10 ºC/min in the programmable electrical furnace. The characterization of sintered samples were done namely: measurement of bulk density, firing shrinkage, vickers hardness and crystal structure by using X-Ray Diffractometer.

3. Results and discussion
Figure 1 shows relationship of bulk density and firing shrinkage to sintering temperature with different of percentages of glass beads. The results show that the pellet sintered for 2 hours with 10 % wt.of glass bead has a higher bulk density value compared to the pellet with 5 % wt.of glass bead. This can be explained that sample with the higher of percentage of glass beads will increases the quantity of melts of glass bead and can enter between the grain so as to cover the existing cavities, consequently the density tends to increase. Likewise with increasing sintering temperature, the value of bulk density tends to increase. It is seen in Figure 1 that the rate of increase in density from 1300 ºC to 1400 ºC tends to be constant for sample with 10 % glass beads, indicating that the sintering process has ended. But it is different in the sample with 5% glass beads, where the bulk density value is still increasing with increasing sintering temperature. This indicates that the sintering process is still continuing at higher temperatures.
Figure 1. The curve of relationship between bulk density and temperature sintering with different composition of glass beads

The highest density of 3.68 g/cm³ was achieved in samples with 10% glass beads and at sintering temperature of 1400°C. When compared with theoretical density value of Al₂O₃ that is 3.95 g/cm³ [11] to the experimental results, it shows that samples with additive 10% glass beads have a density level of 93.16% with a sintering temperature of 1400°C. These results indicate that the glass beads can accelerate and promotes the densification during sintering stage.

Figure 2 shows relationship between firing shrinkage and temperature sintering with different composition additive of glass beads.

Figure 2. The curve of relationship between firing shrinkage and sintering temperature with different composition of glass beads.

Figure 2 shows that the firing shrinkage for both samples increase as the firing temperature is increased, the firing shrinkage value of sample with 10% glass beads is higher than sample with 5% glass beads. The optimum of sintering process is achieved when bulk density and firing shrinkage are a maximum value, tending to be constant. The results of measurement of vickers hardness can be seen at figure 3. Sample after sintering below 1250°C cannot be measured because the surface of the samples was still roughness and contains more pores. It is seen that the Vickers hardness increases with sintering temperatures and composition of glass beads.
Figure 3. The curve of relationship between vickers hardness and temperature sintering with different of composition of glass beads.

The hardness value of sample with 10% glass beads is higher than sample with 5% glass beads, it is due to densification of samples, where sample with 10% glass beads has highest density value. The maximum value of vickers hardness is achieved about 780.4 Hv (7.65GPa), theoretically vickers hardness of alumina ceramic (99.0% Al$_2$O$_3$) is about 10 GPa [11]. Figure 4 shows the results of XRD measurement for sample after sintering at temperature 1400 °C with different composition of glass beads.

Figure 4. XRD patterns of sintered at 1400°C samples with 5% and 10% of glass beads

The analysis the crystal structure of sintered samples were conducted by using X-ray Diffractometer-Rigaku. Based on the matching of the diffractogram peaks with JCPDS card10-173
(corundum) and JCPDS card 15-776 (mullite), the XRD peaks as shown in figure 4, have two phases: corundum (α-Al₂O₃) as major phase and mullite (3Al₂O₃.2SiO₂) as minor phase. The formation of mullite phase is due to reaction between Al₂O₃ with glass beads which glass beads contains a silica or SiO₂.

4. Conclusions
The use of glass beads additives containing the main components of silica can have a significant effect on the decreasing sintering temperature of alumina ceramic. Alumina ceramics can be successfully sintered with a densification level of 93.16 % at sintering temperature 1400°C and with the addition of 10 % wt.of glass beads additives. The properties of ceramic alumina at this condition are bulk density value = 3.68 g/cm³, firing shrinkage value = 15.23 %, vickers hardness value = 780.4 Hv (7.65 GPa). The sintered 1400°C of ceramic alumina with additive 5 % and 10 % of glass beads contain two phases which are corundum as major phase and mullite as minor phase.

Acknowledgments
The authors would like to thank to operators of Vickers hardness and XRD at the Research Center for Physics-LIPI.

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