Effect of Addition of Amorphous Glass (Soda Lime Glass) on Sintering Process and Properties of Alumina Ceramics

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Abstract. The composition of amorphous glass was varied such as 0, 5 and 10 % wt. also temperature of sintering was varied from 1200 to 1400 °C. The γ-Al$_2$O$_3$ powder (p.a) and amorphous glass from soda lime glass were mixed and milled by using a ball mill for 24 hours, and then dried at 100 deg.C. Then, the dried powders were mixed with 2 % PVA as a binder, then, the powders were put into a dies mold and pressed with a pressure of 40 MPa. The pellet samples were sintered by using electrical furnace Thermolyne with heating rate 10°C/minute and holding time at each sintering temperature for 2 hours. The characterization of the sintered sample was done such as measurement of density, porosity, vickers hardness, and analysis of crystal structure. The characterization results show that the highest density value of 3.36 g/cu.cm and the lowest porosity value of 1.06 % obtained at 1400 °C sintering temperature with 10 % additive. While the samples without additive have density value about 2.37 g/cu.cm and its porosity value about 20% at 1400 °C sintering temperature. Similarly, the highest hardness value was achieved at 775 Hv on samples with 10% additive and sintering temperature of 1400 °C. The XRD analysis showed that samples with SiO$_2$ additive up to 10% did not cause reaction and did not cause new phase. The phase that appears only the corundum phase ($\alpha$-Al$_2$O$_3$). The sintering process of Alumina Ceramic the presence of SiO$_2$ additive can be accelerated with lower temperatures.

1. Introduction
Polymorphic crystalline aluminium oxides with the chemical formula Al$_2$O$_3$, and commonly referred with the generic name of alumina, are extensively used in numerous industrial applications (ceramics, electronic substrate, abrasive materials, absorbents, catalysts, biomaterials, composites, pigments, etc) [1]. Among the various structural forms, only the alpha alumina ($\alpha$-Al$_2$O$_3$), or corundum is the thermodynamically stable [1,2]. Densification 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 compaction route for the variety applications must undergo this sintering steps [3]. Sintering is a process where the fully densification will takes place and involves the development of new ceramic structure [3,4]. Typically, this process need to be carried out at nearly melting...
temperature of ceramic. This mechanism of sintering occurred by diffusion transport of matter along definite path which commonly occur at the boundary of stacked or closed particle. The diffusion process that involves during sintering will lead to the interaction between pores and grain boundaries and finally leads to movement of grain growth and the development of a new dense grain structure which preferably in the polycrystalline form [1,5]. Hence, this will increase the strength of the sintered powder due to the bonding and growth of necks between the particles [3] Basically at this stage also, the particles are coalesced (including grain and pore growth) by solid state diffusion with at the same time both of the grain growth and densification occur simultaneously [3,5,6]. In ceramic materials, grain growth can be described by normal and abnormal grain growth [3-5]. So, the controlling the abnormal grain growth is important in achieving a homogenous and higher density. Basically, the fabrication of the ceramic with high density and controlled grain size is depending on reducing grain growth rate and increasing densification rate or some combination of these two. Some modifications of the fabrication approaches that satisfy one or both these condition include the use of additive (dopant) in order get the desired [7,8]. In fact, the use of additive offers a very effective approach for fabricating a pressureless sintering ceramic with highly density and grain size structure [9]. In this work, a small amount of additive was doped into Al₂O₃ compacted pellets as a sintering aid in order to get a desired effect. Basically, an additive such as MgO, TiO₂, ZrO₂, Y₂O₃, SiO₂ and Li₂O are commonly used in ceramic system to influence the densification process either by reducing sintering temperature and time or suppressing and promoting grain growth or enhancing physical and mechanical properties [8]. In high-alumina ceramic products, the sintering process is the most important and expensive step of the fabrication process. It is well known that the addition of small amounts of certain to non-reactive alumina powders can decrease the sintering/densification temperature of alumina bodies from ~1600 to ~1400°C, largely reducing the energy costs of the process [7,9]. It is to be noted that pure alumina can be sintered to only less than 90% relative density at 1600°C, Lee et al. got 99% of relative density by hot pressing for 5% TiO₂ added sample [9]. Kazumasa T et all reported that sintering alumina ceramic at temperature 1700°C for 20 hours can achieved density value about 3.90 g/cm³ or 100% densification[10]. A glass is an amorphous solid that has been quenched from the liquid state, an amorphous glass can be obtained from various types of glass, including: boron silicate glass, soda lime glass and lead glass. Soda lime glass or soda-lime-silicate glass is one of the most commonly used glass compositions can be found in a wide range of glass products, for example as glass containing and glass windows. Its mean chemical composition comprises: 72.2 % SiO₂; 15 % Na₂O; 6.7 % CaO; 4 % MgO; 1.9 % Al₂O₃ and 0.2 % of impurities [11, 12]. A common silica-soda lime glass (glass windows) is used in this study. Therefore in this work, amorphous-SiO₂ was selected as a sintering aid for the pressureless sintering in densification the Alumina Ceramic. In the present work also, the effect of amorphous - SiO₂ with different composition to sintering process of Alumina Ceramic and their properties were investigated.

2. Experimental Works

The starting material was a high-purity (>99.99%) γ - alumina powder and amorphous-SiO₂ from soda lime glass (window glass) as additive. The additive composition expressed in weight percent was 0, 5 % and 10 %. Both of raw materials were milled by ball milling for 24 hours with using pure Al₂O₃ 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 powders were measured particle size by using Laser Particle Size meter. The fine powder was mixed with 2 % wt. of binder polyvinyl alcohol (PVA) and compacted to form a pellet with 12 mm in diameter by pressing under pressure 40 MPa using stainless steel die. The pellets were then dried for 24 hours under room temperature and sintering were conducted at variation temperature such as: 1200, 1300, and 1400°C for 2 hours with holding time and heating rate of 10 °C/min in the programmable electrical furnace. The bulk density, porosity, vickers hardness and crystal structure of the sintered samples were measured.
3. Results and Discussion
The particle size distribution curve of sample without and with additive amorphous glass is shown in figure 1. The samples powder after milling 24 hours by using ball mill have mean diameter particle about 15.41µm for sample without additive and have mean diameter particle about 15.45µm for sample with additive amorphous glass. This particle size is enough as starting materials for making of ceramic materials.

![Figure 1. Particle size distribution of sample (a) without additive and (b) with additive amorphous glass](image)

Figure 2 shows relationship of bulk density and porosity to sintering temperature with different of percentages of amorphous glass. The density values tend to increase with the increase in the composition of amorphous glass and the increase in sintering temperature, this is because an additive glass will melt at high temperature and cover the void cavity of the grain and according to the mechanism of the sintering process there is also a compaction process and reduce pores. But the porosity values tend to decrease with the increase in the composition of amorphous glass and the increase in sintering temperature.

![Figure 2. Relationship of bulk density and porosity to sintering temperature with different of percentages of amorphous glass](image)

The highest value of density is achieved about 3.36 g/cm$^3$ at sample with 10 % of amorphous glass and sintering temperature at 1400°C. But the lowest of porosity is achieved about 1.06 % at similar additive and sintering temperature. According the litterature that theoretical density value of Al$_2$O$_3$ is...
Based on the results that when compared between the densities obtained from experiments with the theoretical density of alumina, the densification value achieved is around 85%. But it is different in the sample without additive amorphous glass, where the bulk density value is still increasing with increasing sintering temperature. This indicates that the sintering process is still continuing at higher temperatures. The measurement of vickers hardness was conducted for all samples after sintering at temperature 1200 – 1400°C by using Micro hardness tester with load 400 gf. The results of measurement of vickers hardness can be seen at figure 3.

The hardness vickers values tend to increase with the increase in the composition of amorphous glass and the increase in sintering temperature. The highest hardness value was reached around 764-775 kgf/mm² in the sample with 10% amorphous glass additive with a sintering temperature of 1400°C, this is in accordance with the level of sample density, where at a temperature of 1400°C the highest density was reached. But when compared with the hardness of alumina ceramics with a densification level of 100%, it has a hardness of around 1100 - 1200 kgf /mm², so the experiment results achieved with 85% densification level, the hardness achieved is still around 764-775 kgf /mm². The value of hardness from ceramic samples without additives showed that it was still low, and the value still tended to increase steadily with increasing sintering temperature, because at sintering temperature of 1400°C it showed that the densification value was still very low. The analysis the crystal structure of sintered samples with 10% of amorphous glass were conducted by using X-ray Diffractometer-Rigaku. Figure 4 shows the results of XRD measurement for sample after sintering at temperature 1300°C and 1400 °C with 10 % of amorphous glass. XRD pattern of sample sintered at 1300°C is similar with xrd pattern of sample sintered at 1400°C. The only difference lies in the peak height, the higher the sintering temperature, the intensity tends to increase, but in this case there is no change in the crystal structure. Based on the matching of the diffractogram peaks with JCPDS card1-089-7117 (corundum) show that the XRD peaks as shown in figure 4. have one phases : corundum (α-Al₂O₃) as dominant phase, There are not the formation of others phase. So in this case the addition of amorphous glass to 10% by weight does not cause the formation of a new phase other than corundum. So the main role of adding amorphous glass additives is only to accelerate the sintering process and increase the densification level.
Figure 4. XRD patterns of samples with 10% of amorphous glass sintered at 1300°C and 1400°C.

The Scanning Electron Microscopy (SEM) was done to analyze the microstructure of sample with 10% amorphous glass and sintered at 1300°C and 1400°C. The SEM micrographs of these specimens are shown in Figure 5.

Figure 5. SEM micrographs of sample with 10% amorphous glass and sintered at 1300°C and 1400°C.

Based on the photos SEM in figure 5 show that there is an enlargement of the grain (size such as 1 µm to 4 µm) from the sample sintered at 1300°C to the sample sintered at 1400°C. Similarly, there is a reduction in pore size, so the microstructure photo of the sample being sintered at 1400°C is more denser.

4. Conclusions
The effect of adding amorphous glass on the sintering process of alumina ceramics can accelerate the sintering process and increase the level of densification. With the addition of additives as much as 10% by weight of amorphous glass and sintering temperature of 1400°C can reach a level of densification of 85%. The properties of ceramic alumina at this condition are bulk density value = 3.36 g/cm³, porosity value = 1.06 %, vickers hardness value = 775 Hv. According XRD results, the sintered 1400°C of ceramic alumina with additive 10% of amorphous glass contain single phases such as: corundum phase (α-Al₂O₃) and it has more denser structure according SEM results.
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