

## Analysis of Particles Size Distribution on the Agglomeration and Shrinkage of Alumina-Zirconia Compacts

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### ABSTRACT

*The combination of Alumina and Zirconia has emerged as a promising ceramic structure for advance machine tool application. However, the particles of Alumina and Zirconia tend to agglomerate during mixture which affected shrinkage and dimension accuracy of the end product. This study focused on the analysis of the particle size of Alumina-Zirconia compacts and their relationship with the shrinkage and agglomerates. The particles size of single Alumina, Zirconia and ball-milled Alumina-Zirconia with 90-10 wt% ratio were examined by mastersizer. These powders then were compacted and sintered at 1400°C to examine their shrinkage. The results show that Alumina possesses larger particles size of 109.65 µm, which is 10 folds larger than Zirconia at 6.10 µm. When blended by ball mill, the Alumina-Zirconia particles were changed into 9.77 µm, showing that the ball mill to refine powder particles while reducing the risk of agglomeration. After sintering, the Alumina-Zirconia compacts were shrunk to maximum 9.56% when 75-25 wt% of Alumina-Zirconia. The combination of porosity, agglomerate and infiltration of zirconia between alumina grains were responsible for the shrinkage of Alumina-Zirconia compacts.*

**Keywords:** Alumina, Zirconia, Particle Size. Agglomerate, Shrinkage.

### 1. INTRODUCTION

Engineering ceramics is the mixture of ceramic powders manufactured for specific function with a controlled process. Generally, engineering ceramics possess high hardness, refractory, chemical inertness and stable at high temperature [1]. The characteristics of engineering ceramics are different depending on the application whether in automotive, aerospace, oil and gas, medical and electronic industries. To name a few, some potential ceramic powders that can be processed as engineering ceramic are Alumina, Zirconia, AlN, SiC, Si<sub>3</sub>N<sub>4</sub>, Ferrites and Piezoceramics [2-3].

One of the engineering ceramic that gains interest from many researchers is Alumina-Zirconia or sometimes called Zirconia Toughened Alumina. Alumina-Zirconia consists of Alumina and Zirconia blended using a powder metallurgy process. The nature of the two materials enables the production of a compact and sturdy structure. The use of Alumina powder in the fabrication of ceramic cutting tools is sufficient to produce a strong structure and hardness. However, the resulting body is fragile and very sensitive to thermal shocks and surprise power [4]. In high-speed machining, tooling is always exposed to a sudden increase in temperature and workload. The risks of failure include cracking, thus causing alumina inefficient use of tooling for high-performance operations.

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On the other hand, the addition of Zirconia around 20% to 25%, can improve some of the weaknesses in the Alumina structure. According to Iftikhar *et al.* [5], high Zirconia content in an Alumina structure increases the fracture resistance up to 50%. Zirconia also acts as a barrier to the increase in Alumina grain size during the sintering process, so that the grain size on the cutting tool will be not too large as larger grain size can cause more material loss when slippage occurred at the grain boundary. With the addition of Zirconia, cutting tools with a density of 99% can be produced at low sintering temperatures [6].

The compaction and density of Alumina-Zirconia were strongly influenced by the particles size of Alumina and Zirconia. Smaller particle size tends to produce higher density, uniform grain size and more compact particle packing. However, there are some challenging issues when the fine size of powders being used. Small particles size and hence large surface area has a high tendency to produce coarse agglomerate as a result of particles sticking together under van der Waals forces [7-8]. The clotted grain would facilitate inherent porosity and will affect the solidification of grains during sintering. Furthermore, this will alter the shrinkage of Alumina-Zirconia compacts which in the end affected the dimensional accuracy of the end product [9-10]. To produce a high performance cutting tool, the hard agglomerate is unwanted since the agglomeration would lead to the formation of coarser grain and consequently retard the hardness and fracture strength of the sintered samples.

The study regarding agglomeration of ceramic particles have been previously conducted by Kocjan *et al.* [11]. The author investigates the agglomeration characteristic when sintering Zirconia nanoparticles. The authors highlighted the mechanism of agglomeration at the rapid sintering condition in relation to rapid grain growth, coalescences of particles and rapid densification. In another study, Vladimir *et al.* [12] analysed surface diffusion of the large volume of Alumina-Zirconia under rapid sintering condition. The grain size and microstructure of Alumina-Zirconia under different sintering rate were analysed. The authors proposed the best parameters that enable the production of dense Alumina-Zirconia. The effects of the particle size of Zirconia on the properties of Alumina-Zirconia have been studied by Zadorozhnaya *et al.* [13]. The authors presented that densification of Alumina-Zirconia increased the mechanical properties and wear performance of Alumina-Zirconia compacts. The author suggested that the grain growth of Alumina during sintering inhibited by the presence of Zirconia particles to facilitate uniform grain size.

Although many studies in the literature have served the understanding of ceramic particles in relation with sintering conditions and grains size distribution, a general relationship between the primary particle size, agglomeration and shrinkage of the ceramic compact are still limited. Hence, the present study investigates the relation between particles size and surface area on the shrinkage of Alumina-Zirconia compact and their relationship with agglomerate. The Alumina-Zirconia is then compacted under uniaxial conditions and sintered in the form of a cutting tool. This study aims to illustrate the effect of primary particle size and to develop an improved understanding of the agglomerate formation that make submicron powders so difficult to compact.

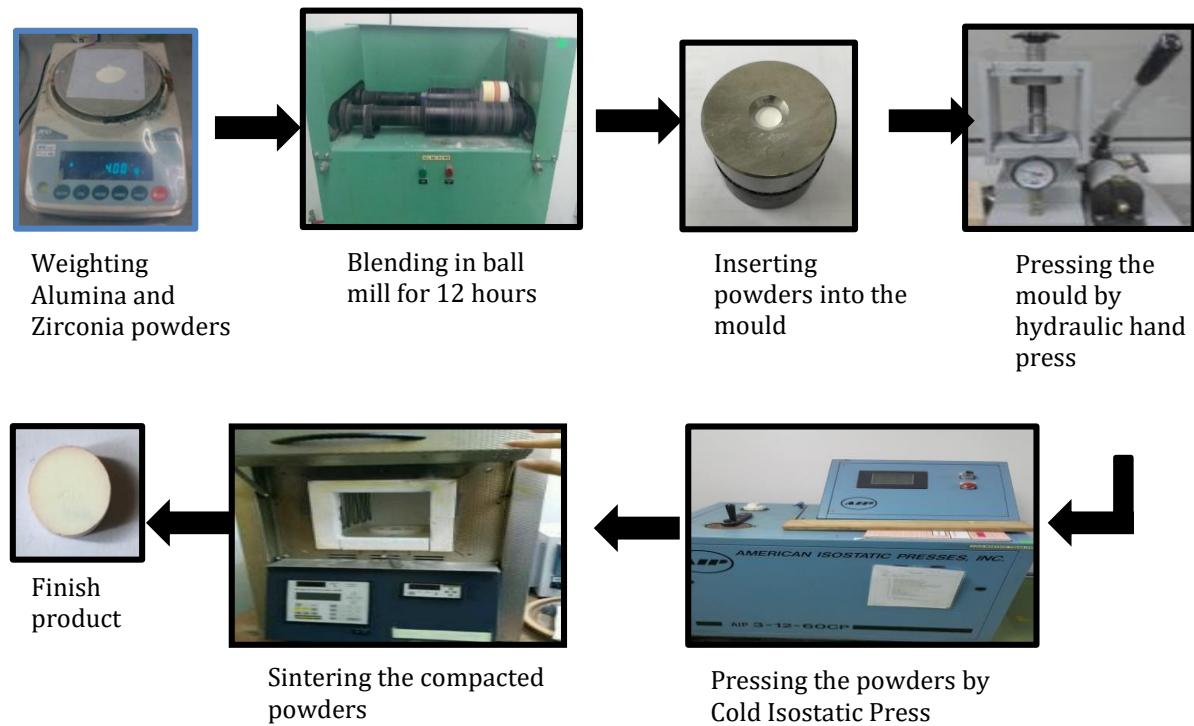
## 2. MATERIAL AND METHODS

The experiments were accomplished with two different stages. The first stage focused on the determination of particles size and surface area of each powder used in this study. By means, powders of Alumina, Zirconia and mixing of Alumina-Zirconia with 90-10 wt% ratio were examined inside Malvern Mastersizer as shown in Figure 1 to determine the particle size distribution and surface area.

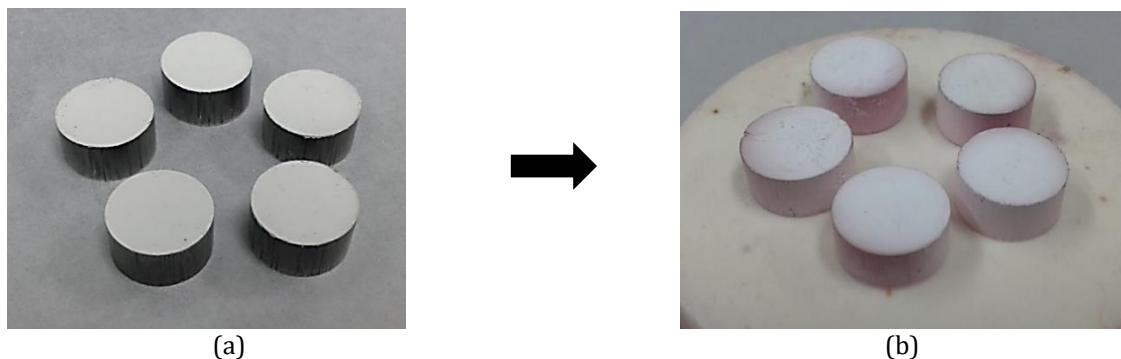
Powders of Alumina and Zirconia with the ratio 90-10 wt% were dry-mixed with the addition of PEG binder to obtain platelets reinforcement. This mixture was ball milled up to 12 hours with 40 rpm to provide homogeneity throughout the sample. This compound was subsequently inserted into the mould and pressed using manual hydraulic press utilizing 5 ton die pressing in the form of RNGN 120600 cutting tool. This is followed by Cold Isostatic press at 350 MPa to further compact the insert. Pressureless sintering was performed at a constant 1400°C and 9 hours soaking time. Pre and post-sintering dimension analysis were applied to some samples to investigate the shrinkage of the ceramic compact. Figure 2 shows the sequence of the compaction of the powder for shrinkage analysis. Figure 3 shows the Alumina-Zirconia compact before and after sintering.



**Figure 1.** Malvern Mastersizer.



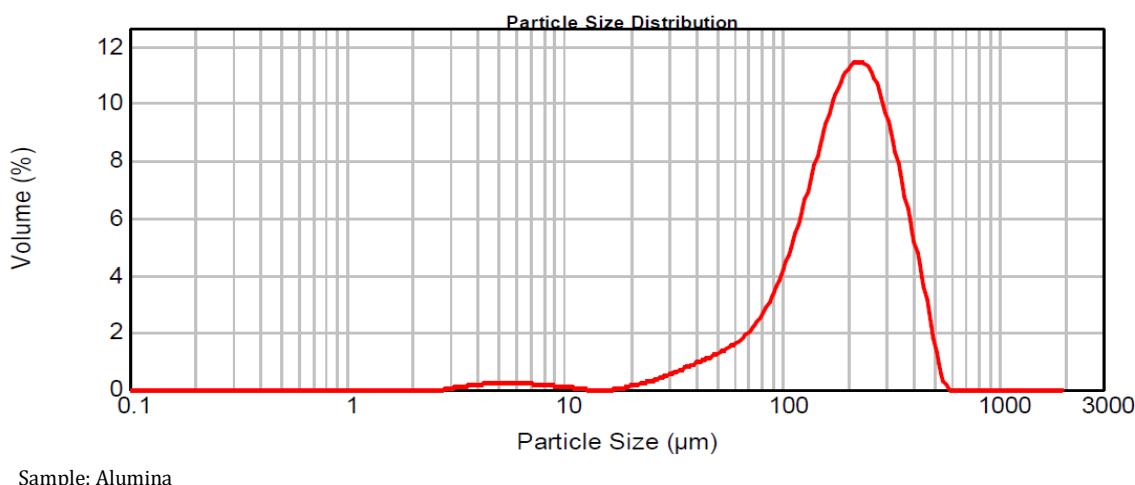
**Figure 2.** The procedure to fabricate Alumina-Zirconia cutting tool.



**Figure 3.** Compacted sample (a) before sintering and (b) after sintering.

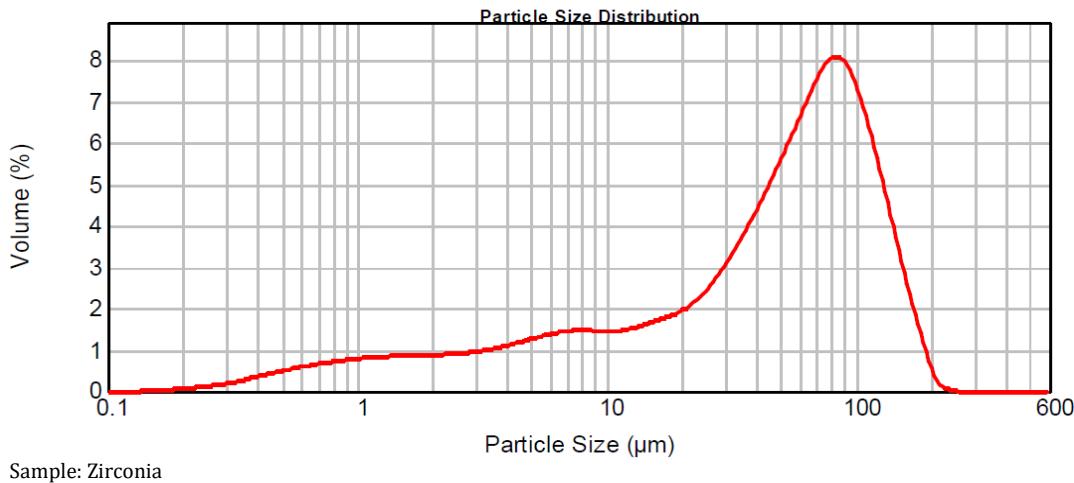
### 3. RESULTS AND DISCUSSION

Early assessment was focused on measurements to determine the particle size of powder Alumina, Zirconia and Alumina-Zirconia used to develop the ceramic cutting tools through particle size analyser equipment. Figure 4 shows the particle size distribution of Alumina powder. The value obtained for the specific surface area of Alumina was  $0.0547 \text{ m}^2/\text{g}$  with an average particle size of  $109.646 \mu\text{m}$ .



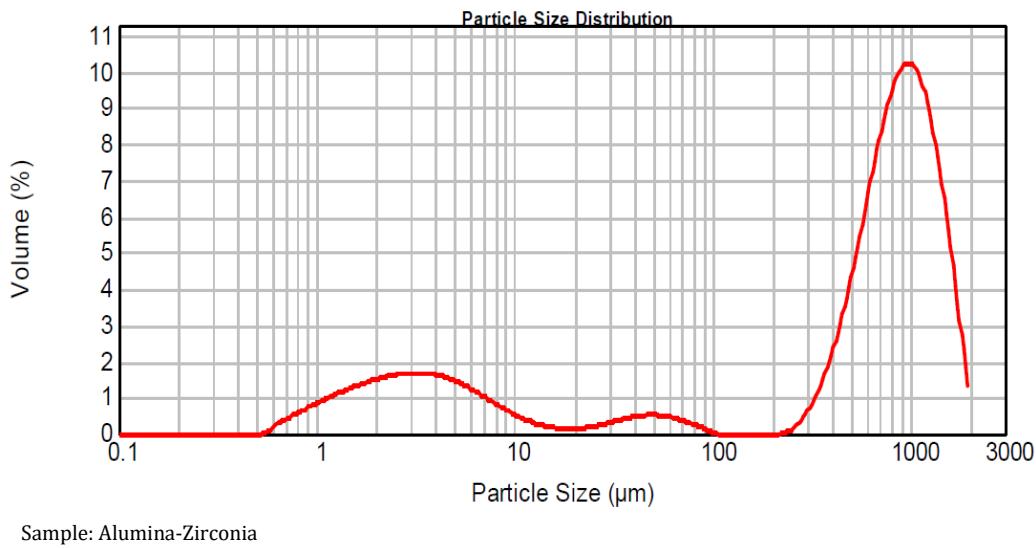
**Figure 4.** Particle size distribution of Alumina powder.

Figure 5 shows the average particle size distribution of Zirconia powder. The specific surface area of Zirconia particle measured at  $0.983 \text{ m}^2/\text{g}$ , whereas the average particle size measured was  $6.103 \mu\text{m}$ . This shows that Zirconia has a smaller average particle size and higher surface area than Alumina. Noted that the average particle size of Zirconia was a lot smaller than Alumina particles. A smaller particle has a bigger surface area and increased the possibility of agglomeration.



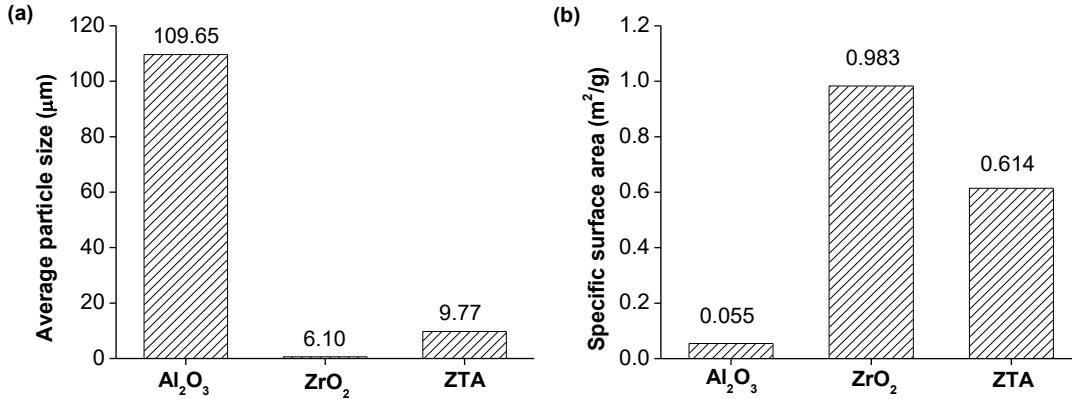
**Figure 5.** Particle size distribution of Zirconia powder.

Figure 6 shows the distribution of particle size for Alumina-Zirconia powders which consist of the 80 wt% Alumina and 20 wt% Zirconia mixture that has been ground through ball mill for 12 hours. The milling process gives an impact on the average particle size and surface area. Noted that, the average particle size for Alumina-Zirconia was 9.77  $\mu\text{m}$  with a specific surface area of 0.614  $\text{m}^2/\text{g}$ . However, there some evident powder agglomeration due to the existence of the very fine particle.



**Figure 6.** Particle size distribution of Alumina-Zirconia powder.

Figure 7 shows that Alumina has a larger particle than Zirconia and Alumina-Zirconia. However, the surface area of Alumina is smaller than Zirconia and volume, v, which reduces the possibility of agglomeration in Alumina. On the observation, the Zirconia has the highest surface area due to the existence of a finer particle which consequently increases the risk of agglomeration of powder. However, when the Alumina and Zirconia powders were ground, powder has smaller particle sizes than Alumina and lower surface area than Zirconia. This proves the effectiveness of the ball mill to refine powder particles while reducing the risk of agglomeration in Alumina-Zirconia powder.



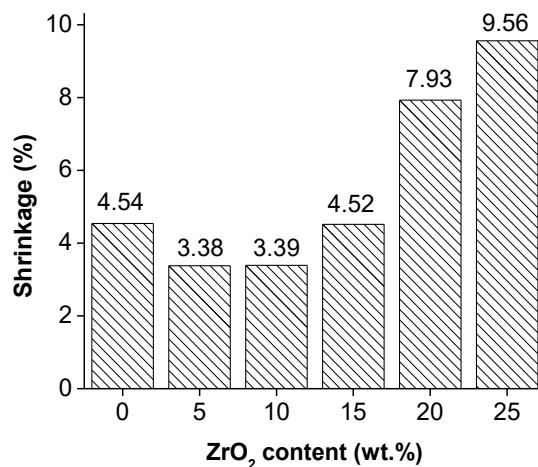
**Figure 7.** The (a) average particle size and (b) specific surface area of each sample.

Agglomeration of ceramic powder is a result of accumulation of fine powder to form a bigger clod of powder. This is because fine powder has the emission of low surface energy from the small specific surface area to attract other particles. Therefore, the attraction between the surfaces of the fine powder is stronger than the gravitational pull mass of the powder and namely known as van der Waals' forces [14]. Agglomeration was influenced by the surface contact area between particles. In theory, the larger surface area increases the attraction between the particles due to the stronger Van Der Waals forces [15]. As shown in Figure 6, Zirconia has the largest surface contact area of  $0.983 \text{ m}^2/\text{g}$ , which means Zirconia has strongest tendency to form agglomerate as compared to Alumina which has the lowest surface area of  $0.0547 \text{ m}^2/\text{g}$ . As for Alumina-Zirconia powder, the surface area is  $0.614 \text{ m}^2/\text{g}$  almost equivalent to Zirconia indicating that the tendency of agglomeration in Alumina-Zirconia is similar to Zirconia. Despite that, it was observed that the particle size of Alumina-Zirconia is lower than Alumina which helps to produce a finer grain structure of the sintered sample.

There are two types of agglomerate which are soft agglomerate, which is easy to be removed and hard agglomerate, which is difficult to be removed. To produce a high-performance cutting tool, the hard agglomerate is unwanted since the agglomeration would result in inhomogeneous distribution between primary and secondary materials. This will prevent the Zirconia particles from reacting actively with the particles of Alumina due to reduction of effective Zirconia content. Another disadvantage of the agglomerate is the outcome product could present undesirable properties related to particles compaction such as porosity, cracks and seizure.

The possibility of agglomerates formation can be reduced by grinding and milling method. In this study, the application of ball mill is one of the grinding method used to refine powder particles as well as avoiding agglomeration. Ball mill process helps to grind and crush the ceramic powder while mixing it evenly [16-17]. This mixing mechanism includes the shearing mixing result from the velocity distribution of powder migration, the friction between the ceramic powder and the impact between particles and powder splits as a result of compression or tensile, generated in the zone between the inner wall and the edge of the stirring bar or bottle [17-18]. The rotational impact forms the powders mixing and homogeneously blended the particles in the rotary tumbler to produce uniform particles distribution. Evidently, the particle size of Alumina has been reduced after ball mill process from  $109.646 \mu\text{m}$  to  $9.773 \mu\text{m}$  indicating removal of soft agglomerate present in Alumina-Zirconia powder.

Figure 8 shows the effect of Zirconia addition on the shrinkage for Alumina-Zirconia compacts. The graph shows that shrinkage percentage increase with increasing of Zirconia weight percentage in Alumina matrix. The largest shrinkage recorded at 9.56% when 25 wt% Zirconia was added into the Alumina matrix.



**Figure 8.** Graf of shrinkage for each composition.

The shrinkage of ceramic particles after sintering is expected due to the different thermal expansion of Alumina and Zirconia. According to Azhar *et al.* [9], Zirconia has a larger thermal expansion of  $8.1 \times 10^{-6} \text{ }^{\circ}\text{C}^{-1}$  as compared to Alumina,  $10.3 \times 10^{-6} \text{ }^{\circ}\text{C}^{-1}$ . In the structure that contained a large size of Alumina particles and smaller size of Zirconia particles, the large thermal expansion of Zirconia particle in the Alumina has reduced the space for Alumina grain to grow. At the same time, high sintering temperature increased liquidity of particles which forced them into diffusion when engaged each other [10]. Once diffused, the separation distance of the mass centre between particles would also reduce, which eventually cause shrinkage along with the sintered particles. Overall, shrinkage in this study only consumed up to 9.56%. Prediction of the shrinkage characteristics of ceramic composites is important to produce sintered ceramic with grain anisotropy, defect-free and near net shape [19-20].

#### 4. CONCLUSION

This study focused on the analysis of the particle size of Alumina-Zirconia compacts and their relationship with the shrinkage and agglomerates.

- i. The particle size of Alumina powder has an average particle size of 109.65  $\mu\text{m}$  which is 10 folds higher than Zirconia of 6.10  $\mu\text{m}$ . When the powders are mixed (Alumina-Zirconia) and ball milled for 12 hours, the total particle size obtained is 9.77  $\mu\text{m}$ . Smaller particle size provided better densification and resistance to deformation.
- ii. The highest surface area was recorded by Zirconia at 0.983  $\text{m}^2/\text{g}$  and followed by Alumina at 0.0574  $\text{m}^2/\text{g}$ . Higher surface area promotes more tendency to form an agglomerate. Agglomerate is unwanted due to inhomogeneity issue and disturbance of particle packing.
- iii. When sintering single Alumina, abnormal grain growth was produced where some particles expanded larger than others. When Zirconia was added into Alumina matrix, the grain growth of Alumina can be controlled into smaller and uniform size due to infiltration of small Zirconia between particles of Alumina.
- iv. Sample with 25 wt% of Zirconia content was the most consistent and closed packed showing highest shrinkage, reflected particle size mismatch and grain diffusion during sintering.

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