Investigation of mechanical properties based on grain growth and microstructure evolution of alumina ceramics during two step sintering process

U. A. Khan, A. Hussain, M. Shah, M. Shuaib and F. Qayyum
Department of Mechanical Engineering, University of Engineering and Technology, Taxila 47050 Pakistan
†Ibn-e-Seena Institute of Technology, Sector H-11, Islamabad, Pakistan
E-mail: umar.ali.2289@gmail.com

Abstract. Alumina ceramics having small grain size and high density yield good mechanical properties, which are required in most mechanical applications. Two Step Sintering (TSS) is used to develop dense alumina ceramics. In this research work the effect of sintering temperatures on microstructure and density of the alumina specimens developed by using TSS has been investigated. It has been observed that TSS is more efficient in controlling grain growth and increasing the density as compared to One Step Sintering (OSS) of alumina. Scanning electron micrographs of sintered alumina specimens have been compared. It has been observed that TSS proves to be a better technique for increasing density and controlling grain growth of alumina ceramics than OSS. More relative density, hardness, fracture toughness and small grain size was achieved by using TSS over OSS technique.

1. Introduction
Alumina ceramics are well known and widely used as engineering material due to their good mechanical properties like hardness, wear resistance, and structural stability at elevated temperature. They are widely used for making good quality abrasive tools, seal rings, thread guides, cutting tools etc. It has been continuously found by different researchers that alumina ceramics having small grains and fine microstructure show improved strength [1], hardness [2] and wear resistance [3].

Sintering process involves coalescence of powder particles into dense object under action of heat in which accelerated grain growth occurs at final stage of sintering [4]. Since the invention of process many researcher have devised several sintering techniques to attain required density and grain size which eventually helps in achieving required mechanical characteristics. The techniques involved are Pressureless sintering and Pressure assisted sintering. Pressureless sintering involves sintering of powder compact without applying pressure during heating that is economical and produces near net shaped parts [5-7]. High temperature is required to achieve good densification which also increases grain size. Pressure assisted sintering utilize pressure on powder compact during heating which is important for manufacturing dense parts of ceramics quickly [4]. Spark Plasma sintering [2, 8, 9], hot pressing sintering [3, 10], and hot Isostatic pressing [1, 11]are a few well know pressure assisted sintering techniques, these techniques help in reduction of grain growth speed while achieving high density.

Two step sintering (TSS) is a Pressureless sintering technique which was developed by Chen and Wang [12] in year 2000 to reduce the grain growth in yttria ceramic in the final stage of sintering.
Later on many researchers have successfully used TSS to increase ceramic density and to control grain size during sintering of Yttria [12, 13], Barium Titanate [14], Zinc Oxide [15, 16], Alumina [15, 17, 18] and Silicon carbide [11] specimens.

In current research alumina ceramic was manufactured by using OSS and TSS techniques, the effects of sintering temperature on microstructure and density of the specimens were observed. Mechanical properties on the basis of grain growth and microstructure evolution were presented. The etched specimens were observed under TESCAN Field Emission Scanning Electron Microscope (FESEM) to determine their grain sizes and the results have been presented.

2. Experimentation
Alumina powder CT 3000 (Almatis) designated as AP-1 and alumina powder (Nanophase Technology) designated as AP-2 were used for sintering of specimens. Powder specifications are mentioned in Table 1.

| Alumina Powder | Purity   | Particle Size | Specific Surface Area (m²/g) |
|----------------|----------|---------------|------------------------------|
| AP1            | 99.7%    | 600 nm        | 7                            |
| AP2            | 99.9%    | 47 nm         | 35                           |

Alumina green compacts were made in a custom die (having 28mm internal diameter) mounted on Material Testing System MTS 810 which is shown in Figure 1 having force capacity of 100 kN. Uniaxial pressure of 105 MPa was applied to successfully develop green compacts.

![Figure 1. Test setup for uniaxial pressing](image)

The green density is measured by volumetric method. S-1, S-3 specimens were formed by Alumina Powder AP-1 (600 nm) and S-2, S-4 specimens by alumina powder AP-2 (47nm). The green density and specimens dimensions are shown in Table 2.
Table 2. Green Density of AP-1 & AP-2 Specimens pressed at 105 MPa

| Specimen | Mass (g) | Mass of Al₂O₃ (g) | Diameter (mm) | Thickness (mm) | Green Density (g/cm³) | Relative Density (%) |
|----------|---------|-------------------|---------------|----------------|----------------------|----------------------|
| S-1      | 5       | 4.37              | 28            | 3.40           | 2.08                 | 52.1                 |
| S-3      | 5       | 4.34              | 28            | 3.40           | 2.07                 | 52                   |
| S-2      | 3       | 2.28              | 28            | 2.9            | 1.27                 | 31.8                 |
| S-4      | 3       | 2.16              | 28            | 2.85           | 1.23                 | 30.8                 |

During OSS green compacts were heated to 1450°C for soaking time of 6 hour while during TSS the green compact to 1450°C then suddenly cooled down to 1350°C for soaking time of 6 hour shown in Figure 2. Electric furnace with SiC heating elements (Vecstar) of heating chamber size 5.9×7×13.9 (inch) was used for sintering of green compacts.

![Figure 2. OSS and TSS sintering Curves](image)

Densification of sintered alumina specimens was measured by Archimedes technique using MD-300s balance in which specimen values are measured by immersing them in distilled water. The measurements are presented in Table 3.

Table 3. Bulk density of AP-1 & AP-2 sintered specimens

| Specimen | Diameter (mm) | Thickness (mm) | Bulk Density (g/cm³) | Relative Density (%) |
|----------|---------------|----------------|----------------------|----------------------|
| S-1      | 24            | 3.22           | 3.23                 | 81                   |
| S-3      | 23.30         | 3.07           | 3.61                 | 90.5                 |
| S-2      | 20.80         | 2.38           | 3.56                 | 89.3                 |
| S-4      | 19.72         | 2.29           | 3.66                 | 91.8                 |

As discussed previously that microstructure of alumina ceramics play an important role in defining mechanical properties. To reveal the microstructure of the specimens and prepare them for microscopy, the specimens were ground using 400, 800, 1200 and 2000 grade emery papers consecutively. The specimens were polished with 1 µm diamond paste with the help of a buffing wheel. The specimens were thermally etched at 1250°C for 30 min in electric furnace and observed under TESCAN Field Emission Scanning Electron Microscope (FESEM).

Vickers hardness (Hᵥ) measurements were conducted by a fully calibrated Vickers hardness tester (Time Instrument). Indentations were conducted on polished faces of the specimens with a diamond indenter using 10 kg load for 15 sec. Three measurements were taken and average result are reported using Eq. (1)[19].
\[ H_V = 1.854 \left( \frac{P}{d^2} \right) \]  \hspace{1cm} (1)

Where \( P \) is applied load in Kgf and \( d \) (mm) is mean value of diagonal length.

Fracture toughness \( (K_c) \) was computed by following Eq. (2) [20].

\[ K_c = 0.0726 \left( \frac{P}{c^2} \right) \]  \hspace{1cm} (2)

Where \( P \) is applied load in Kgf and \( c \) (mm) is the mean crack length of indented marks.

3. Results and discussion

Green compacts were manufactured on MTS 810 system, due to its fully calibrated load cells and LVDT’s we were able to record detailed compactness curves of alumina powder which are presented in Figure 3. Compactness curves shows that AP-2powder was compressed more than AP-1 during uniaxial pressing which is due to fine and small particle size of AP-2 powder. The finer alumina powder gives greater contact points of particles which form cold welds in green compact due to which powder compactness increases[21].

Figure 3. Pressure vs displacement curve of alumina powders during uniaxial pressing

The results obtained after OSS and TSS are summarized in Table 4. The results showed that alumina powder AP-2 shrinked more than AP-1 because of fine particles, high specific surface area of the AP-2 powder and greater compactness during uniaxial pressing. Fine particles have shorter diffusion distances and larger area of diffusion. High surface area of powder delivers higher driving force and stimulates densification leading to decrease of interfacial energy [22].

Also TSS has increased specimens relative density of AP-1 specimens by 9.5 % and AP-2specimens by 2.5% as compared to OSS specimens. The microstructure of alumina specimens obtained by one step sintering and two step sintering are shown in Figure 4 and 5. It was observed that two step sintered specimens have small grains as compared to one step sintered and both contain inter granular porosity. During sintering of ceramics open pores stick grain boundaries and block grain boundary motion at intermediate stage but when open pores close, grain growth starts to occurs[23]. Chen and Wang developed two step sintering, to increase density without grain growth, grain boundary diffusion must be active and grain boundary migration must be frozen [12]. They found that grain boundary migration require higher activation energy than grain boundary diffusion so they decreased temperature in second step (T2) in order to freeze grain boundary migration.

It can be seen from SEM micrographs that TSS specimens S-3 and S-4 have grain sizes of 1.28 \( \mu \)m and 0.76 \( \mu \)m which is smaller than the grain size of OSS specimens.
### Table 4. Green and Bulk density of sintered specimens

| Specimen | Green Density (g/cm³) | Bulk Density (g/cm³) | Densification (%) |
|----------|-----------------------|----------------------|-------------------|
| S-1      | 2.08                  | 3.23                 | 35.6              |
| S-3      | 2.07                  | 3.61                 | 42.6              |
| S-2      | 1.27                  | 3.56                 | 64.3              |
| S-4      | 1.23                  | 3.66                 | 66.4              |

![Figure 4](image1.png)

**Figure 4.** SEM micrographs of AP-1 specimens (a) S-1 OSS (b) S-3 TSS

![Figure 5](image2.png)

**Figure 5.** SEM micrographs of AP-2 specimens (a) S-2 OSS (b) S-4 TSS

The variation of relative density and grain size of alumina ceramic specimens sintered via one step and two step sintering are shown in Figure 6. One step sintered specimens S-1 and S-2 have large grain size because the decrease in total interfacial energy arises via densification and particle coarsening, smaller particles merge with the large ones producing larger grains[24]. While Two step sintered...
specimens S-3 and S-4 have small grain size because specimens heated to high temperature to decrease pores that are unstable against shrinkage and cooled at lower temperature to attain densification via suppression of grain boundary migration [12].

![Grain size versus relative density of alumina specimens sintered by single and two step sintering](image)

**Figure 6.** Grain size versus relative density of alumina specimens sintered by single and two step sintering

Mechanical properties of OSS and TSS specimens were calculated by using Eq. (1) [19], and Eq. (2) [20]. are shown in Table 5. As grain size has strong influence on mechanical properties [1] because fine grain size of a microstructure contain high density of grain boundaries that restrain deformation of specimen in loading.

**Table 5. Mechanical Properties of sintered alumina specimens**

| Specimens Sintered | Hardness (GPa) | Fracture Toughness (MPa.m$^{1/2}$) |
|--------------------|---------------|----------------------------------|
| S-1                | 12.41         | 1.53                             |
| S-3                | 13.27         | 1.87                             |
| S-2                | 13.64         | 2.16                             |
| S-4                | 14.45         | 2.30                             |

It is found that TSS has controlled alumina grain growth and increased density, hardness and fracture toughness.

**4. Conclusion**

I. Hence it is evident from research that alumina ceramics developed by using fine powder and TSS technique yield better ceramics of smaller grain size, increase hardness and toughness value.

II. Two step sintering has increased density and controlled grain size of AP-1 (600 nm) and AP-2 (47 nm) ceramics.

III. Alumina AP-2 (47 nm) showed greater sinter ability due to small powder particle size, high compatibility and specific surface area of the powder.

**5. Acknowledgements**

I. The authors are grateful:

II. For financial support from Board of Post Graduate studies, University of Engineering and Technology Taxila, Pakistan

III. For specimen characterization support, Institute of Space Technology and Ibn-e-Seena Institute of Technology Islamabad.
6. References

[1] O, Y.T., et al., Effect of grain size on transmittance and mechanical strength of sintered alumina. Materials Science and Engineering: A, 2004. 374(1–2): p. 191-195

[2] Santanach, J.G., et al., Spark plasma sintering of alumina: Study of parameters, formal sintering analysis and hypotheses on the mechanism(s) involved in densification and grain growth. Acta Materialia, 2011. 59(4): p. 1400-1408

[3] Felten, E.J., Hot-Pressing of Alumina Powders at Low Temperatures. Journal of the American Ceramic Society, 1961. 44(8): p. 381-385

[4] M.N.Rahaman, Ceramic Processing and Sintering. 2nd ed. 2003, New York: Marcel Dekker.

[5] Somiya, S., Handbook of Advanced Ceramics: Materials, Applications, Processing, and Properties. 2013: Academic Press

[6] Eskandari, A., et al., Effect of high energy ball milling on compressibility and sintering behavior of alumina nanoparticles. Ceramics International, 2012. 38(4): p. 2627-2632

[7] Cutler, I.B., et al., Sintering of Alumina at Temperatures of 1400°C. and Below. Journal of the American Ceramic Society, 1957. 40(4): p. 134-139

[8] Kim, B.-N., et al., Spark plasma sintering of transparent alumina. Scripta Materialia, 2007. 57(7): p. 607-610

[9] Shen, Z., et al., Spark plasma sintering of alumina. Journal of the American Ceramic Society, 2002. 85(8): p. 1921-1927

[10] Chang, S., et al., Hot-Pressing of Nano-Size Alumina Powder and the Resulting Mechanical Properties. International Journal of Applied Ceramic Technology, 2004. 1(2): p. 172-179

[11] Besson, J. and M. Abouaf, Grain growth enhancement in alumina during hot isostatic pressing. Acta Metallurgica et Materialia, 1991. 39(10): p. 2225-2234

[12] Chen, I.-W. and X.-H. Wang, Sintering dense nanocrystalline ceramics without final-stage grain growth. Nature, 2000. 404(6774): p. 168-171

[13] Wang, X.H., P.L. Chen, and I.W. Chen, Two-Step Sintering of Ceramics with Constant Grain-Size, I. 12O3. Journal of the American Ceramic society, 2006. 89(2): p. 431-437

[14] Polotai, A., et al., A novel approach to sintering nanocrystalline barium titanate ceramics. Journal of the American Ceramic Society, 2005. 88(11): p. 3008-3012

[15] Maca, K., V. Pouchly, and P. Zalud, Two-Step Sintering of oxide ceramics with various crystal structures. Journal of the European Ceramic Society, 2010. 30(2): p. 583-589

[16] Mazaheri, M., A. Zahedi, and S. Sadrnezhad, Two-step sintering of nanocrystalline ZnO compacts: Effect of temperature on densification and grain growth. Journal of the American Ceramic Society, 2008. 91(1): p. 56-63

[17] Kim, H.S., S.T. Oh, and Y.D. Kim, Effects of the two-step sintering process on the optical transmittance and mechanical strength of polycrystalline alumina ceramics. Ceramics International, 2014. 40(9, Part A): p. 14471-14475.

[18] Wang, C.-J., C.-Y. Huang, and Y.-C. Wu, Two-step sintering of fine alumina–zirconia ceramics. Ceramics International, 2009. 35(4): p. 1467-1472

[19] ASTM Standard Designation C 1327-03. Standard test method for vickers indentation hardness of advanced ceramics. ASTM USA :International

[20] Sergejev, F. and M. Antonov, Comparative study on indentation fracture toughness measurements of cemented carbides. Proc. Estonian Acad. Sci. Eng, 2006. 12(4): p. 388-398

[21] Datta, B.K., Powder Metallurgy: An Advanced Technique of Processing Engineering Materials. 2014: PHI Learning Pvt. Ltd

[22] Bowen, P. and C. Carry, From powders to sintered pieces: forming, transformations and sintering of nanostructured ceramic oxides. Powder Technology, 2002. 128(2–3): p. 248-255

[23] Li, J. and Y. Ye, Densification and Grain Growth of Al2O3 Nanoceramics During Pressureless Sintering. Journal of the American Ceramic Society, 2006. 89(1): p. 139-143

[24] Mayo., M., in Int Mater Rev. 1996. p. 85-115