Precompaction Effects on Density and Mechanical Properties of Al₂O₃ Nanopowder Compacts Fabricated by Magnetic Pulsed Compaction

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In this study, the effects of precompaction on the density, microstructure, mechanical and electrical properties of α-Al₂O₃ bulks fabricated by the combined application of magnetic pulsed compaction (MPC) and a sintering process were reported. The obtained density of the α-Al₂O₃ bulks prepared by the combined processes increased with the increasing MPC pressure and precompaction pressure. The resultant higher hardness and breakdown voltage of the consolidated bulks following combined application of the magnetic pulsed compaction, precompaction and sintering process could be attributed to the homogeneously distributed ultra-fine microstructure than that of general processing, suggesting that the grain growth was remarkably reduced during the MPC processes. [doi:10.2320/matertrans.M2009265]

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1. Introduction

Alumina is a ceramic material which has attracted the attention of scientists as well as researchers throughout the world that have developed techniques to make it suitable for a wide range of applications. Alumina is the most commonly used type of ceramic and is available in purities up to 99.99%. Hence, typical applications of alumina include electronics, electrical insulators, seal faces, valve seats, dosimetric applications in radiotherapy, growth of carbon nanotubes on microsized alumina particles and has promising applications in the automotive industry.

However, difficulties arise during the consolidation process when nanostructured powders are exposed to high temperature to obtain a higher density above 90% resulting in the growth of grains causing the loss of its superior properties. Unfortunately, processing these nanopowders into fully dense, bulk products that retain the original nanoscale grain size has proven to be difficult due to a unique combination of problems¹ such as high surface area, severe interparticle friction, and high level of chemisorbed gases.

To avoid some of the limitations imposed by exaggerated grain growth and related problems, a number of other consolidation methods has been evaluated, such as hot-pressing,³ sintering by microwave radiation,⁴,⁵ spark plasma sintering, etc. However, none of these methods have been very successful in producing fully dense bulk products that retain nanoscale grain size. In our previous study, we proposed the use of magnetic pulsed compaction (MPC) for the effective consolidation of nanopowders.⁶,⁷ The known data for pressing nanosized powders by a high pulsed pressure gives us the opportunity of obtaining extremely dense compacts with a nanostructure and high mechanical properties. It represents a significant challenge for researchers to develop a new consolidation process technique, which is capable of suppressing the grain growth as well as simultaneously achieving a higher density, and to further improve the microstructural properties of the finished product.⁷–⁹ Therefore, the objective of this research is to find a better consolidation process to overcome all these adversities.

In this present study, bulk Al₂O₃ nano-ceramics were prepared by a combination of magnetic pulsed compaction and a sintering process, and the effects of precompaction on the green density, grain size, mechanical and electrical properties of the sintered bulks were investigated.

2. Experimental

The starting powder was α-Al₂O₃ (purity of 99.8%) with the average powder particle size of 50~200nm. 1.5 grams sample of the raw Al₂O₃ powder was loaded into a die and punch unit, having outer and inner diameters of 50 and 15 mm, respectively. The Al₂O₃ powder was formed into the shape of a disk by magnetic pulsed compaction (MPC). A graphite paste was used as a lubricant on the die wall and the bottom punch. The pressure of the magnetic pulsed compaction (MPC) varied from 0.5 to 1.8 GPa at room temperature. In order to improve the density and properties, the starting powder was precompacted in a die under 110 MPa, 220 MPa, and 330 MPa, and then each precompacted sample without separation from the die was MPCed at room temperature. The MPCed bulks were subsequently sintered at 1450°C for 3 hrs in the air atmosphere.

The sintered bodies were used for the Vickers hardness test and breakdown voltage testing after polishing. The apparent density of the bulk was measured by the Archimedes method using water, and the values were averaged. The relative density was calculated assuming a true density of 3.987 g/cm³ for the α-Al₂O₃. A microstructural analysis of both the powder and the sintered bodies was conducted using a scanning electron microscope (SEM) and a transmission electron microscope (TEM) equipped with Energy Dispersive X-ray Spectrometers (EDS). X-ray diffraction (XRD) patterns were obtained at a scanning rate of 4°/min in...
the 2θ range from 10 to 80° using a fully automated X-ray diffractometer with Cu Kα (0.15406 nm) radiation. The fracture surfaces were examined by scanning electron microscopy (SEM) to investigate the fracture model and grain size. The Vickers hardness measurements were performed using a Vickers hardness tester with a Vickers indenter at the applied load of 19.6 N for 10 s.

3. Results and Discussion

The typical morphology of the Al₂O₃ nano-powder used in this research, as observed by TEM, has a size range of approximately 150 to 200 nm with spherical and elliptical shapes. For compaction of the Al₂O₃ nanopowders, a magnetic pulsed compaction process was applied, which transforms the pulsed electric power to a mechanical pulse and concentrates the pulse in the compaction zone. In the present investigation, the action of the magnetic field was enhanced by means of special concentrators and pulsed pressures between 1 to 5 GPa increased during 3 to 300 μ·sec in the compaction zone.

In this study, dynamic compaction by magnetic pulsed compaction (MPC) and precompaction were conducted in order to prepare the Al₂O₃ bulk. Table 1 demonstrates the compaction arrangements, the apparent features and final density of the bulks fabricated by the combination of MPC, precompaction, and a sintering process. The defects, such as cracks and dimples on the surface of bulks, were not observed. The obtained density of the MPCed specimen increased with the increasing MPC pressure. However, the bulk MPCed at 1.8 GPa showed fine cracks on the surface. The pressure required to consolidate the nanopowder is related to the force required to push the particles together. In order to push two particles together, the applied force must be equal to or greater than the resisting force. The ceramic powders are not expected to plastically deform during compaction like metals. In hard ceramics, a plastic deformation of the particles or the formation of a particle-to-particle contact is so difficult that the stored strain energy by the compaction pressure could not be readily relaxed, which results in the formation of cracks in the materials compacted at high pressure.

Table 1 Consolidation conditions and relative densities of sintered bulks by a combination of MPC and precompaction processing.

<table>
<thead>
<tr>
<th>Experimental conditions</th>
<th>Bulk condition</th>
<th>Density (%)</th>
</tr>
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<tbody>
<tr>
<td>Uniaxial static compaction</td>
<td></td>
<td></td>
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<tr>
<td>(110 MPa) + Sintering (1450° C for 3 h)</td>
<td>Good</td>
<td>90.0</td>
</tr>
<tr>
<td>MPC (0.5 GPa) + Sintering (1450° C for 3 h)</td>
<td>Good</td>
<td>90.0</td>
</tr>
<tr>
<td>MPC (1.25 GPa) + Sintering (1450° C for 3 h)</td>
<td>Good</td>
<td>92.0</td>
</tr>
<tr>
<td>MPC (1.8 GPa) + Sintering (1450° C for 3 h)</td>
<td>Fine crack</td>
<td>90.0</td>
</tr>
<tr>
<td>Pre-compaction (110 MPa) + MPC (1.25 GPa) + Sintering (1450° C for 3 h)</td>
<td>Good</td>
<td>93.0</td>
</tr>
<tr>
<td>Pre-compaction (220 MPa) + MPC (1.25 GPa) + Sintering (1450° C for 3 h)</td>
<td>Good</td>
<td>94.5</td>
</tr>
<tr>
<td>Pre-compaction (330 MPa) + MPC (1.25 GPa) + Sintering (1450° C for 3 h)</td>
<td>Good</td>
<td>94.5</td>
</tr>
</tbody>
</table>

Based on this result, it is obvious that the maximum pressure required to consolidate the Al₂O₃ nanopowder without any crack generation on the surface of the bulk was 1.25 GPa. The bulk compacted at 1.25 GPa reached a slightly higher density than the bulk compacted at 0.5 GPa in spite of having the same initial density. Upon pressing at 1.8 GPa, the density decreased to 90% due to the formation of fine cracks on the surface of the MPCed and sintered bulk. In addition, it is clear from the plot that the density also increases with the increasing precompaction pressures from 110 MPa to 220 MPa and then becomes saturated at 94.5% for precompaction pressure beyond 330 MPa. However, this density value is slightly lower than that of commercial Al₂O₃ like 96% in this study. The highest density of 94.5% was achieved in the sample precompacted at 220 MPa, while the density of the sample without precompaction was 92%. The means that precompaction of the powder before MPC improves the final density of the sintered bulk. This may be due to the higher initial packing density introduced by precompaction pressure as well as the higher MPC pressure. The increased green density may be due to better packing associated with small particles filling the voids between the larger ones. Therefore, the pressure may play a role in the initial stage through particle rearrangement and distribution of agglomerates as well as in the late stages of the densification. Additionally, it was reported that a high initial density is not only effective to enhance the subsequent densification during sintering, but also limits the rapid grain growth. Therefore, it is necessary to very carefully control the strain energy during the fabrication of ceramic compacts by the application of a very high pressure, like the MPC.

Figure 1 represents the SEM micrographs of the sintered bulk as a function of the MPC pressure and precompaction. The microstructure of a commercial Al₂O₃ plate (Fig. 1(a)) shows coarse grains and porous structures along the boundaries. The microstructure of the MPCed and sintered bulk exhibits small grains with fine pores. The average grain size measured for the 0.5 GPa and 1.25 GPa MPCed pressures without precompaction was 0.74 μm and 0.9 μm as shown in Fig. 1(b) and 1(c), respectively. Likewise, a more refined microstructure with an average grain size of 0.86μm was found for the bulk MPCed at 1.25 GPa and precompacted at 220 MPa as exhibited in Fig. 1(d). On the contrary, the size range of the commercial Al₂O₃ plate was 5.45 μm which is much higher than the MPCed one. Furthermore, the relative density of the sample precompacted at 220 MPa was the highest at 94.5% and most of the resolved porosity appeared to be isolated at the grain interstices. The micro-structural features as revealed from the FE-SEM analysis thus showed the promise of consolidating of nanopowders while retaining ultra-fine microstructures. Moreover, to distinguish the effect of the compaction on the microstructural characteristics between the uniaxial and MPC pressures, specimens were closely examined by TEM. Figure 2 shows TEM micrographs of the uniaxially compacted specimen (Fig. 2(a)) and MPCed specimen (Fig. 2(b)).

A larger grain size with an average of 820 nm together with a considerable porous structure size within the particles were observed in the case of uniaxial compaction (Fig. 2(a)) compared to the specimen compacted by MPC. The higher
MPC pressure significantly influenced the decreased grain size as evident in Fig. 2(b). Besides, the microstructure indicates a homogeneous dispersion of finer grains varying in size from 380 to 840 nm with an average of 600 nm.

Figure 3 shows the variation in the Vickers hardness as a function of the MPC pressure and pre-compaction condition during consolidation. With the increasing MPC pressure, the hardness of the bulk increases and the value is much higher than that of the uniaxial compaction. In addition, the different porosity sizes in different regions can be another important factor leading to the variation. The homogeneously distributed ultra-fine grains and higher density of the MPCed bulk might be the reasons for the increased hardness. The hardness of the bulk also increases with the increasing precompaction pressure. This suggests that particle rearrangement during the precompaction occurs at the lower pressures.

The improved hardness with the increasing precompaction pressure was possibly associated with the enhanced density of the bulk. These results clearly indicate that the precompaction prior to the MPC and sintering is an efficient approach to promote the density and Vickers hardness. Besides, the structural characteristics, which can be controlled by the consolidation and processings, such as the porosities, internal stress, etc., may play an important role in determining the properties. However, it is ambiguous to distinguish the contribution of the property enhancement
between the grain size effect and other effects such as the pores or strains accompanying the consolidated samples.

The reliability of the consolidated bulk is significantly related to the generation and propagation behavior of the microcracks, and the indentation test becomes one of the most effective and convenient methods to determine the fracture of materials. Figure 4 shows representative SEM micrographs of the cracks formed from the corner of the indenter. Despite the secondary cracking, a long crack of 50 µm formed from each corner of the indenter and easily propagated without tortuously in a sapphire Al₂O₃ plate as shown in Fig. 4(a). However, the sintered specimens after the MPC show a decrease in the crack length (about 40 µm) compared to that of the a sapphire Al₂O₃ plate. The crack length in the bulk MPCed at 0.5 MPa was about 50 µm and for the specimen MPCed at 1.25 GPa, it was 40 µm. On the contrary, a crack deflection was observed in Fig. 4(e) and (f), as a result of the uniformly dispersed fine particles in the MPCed and sintered bulk. The interaction between the

![Fig. 3 Vickers hardness of sintered bulks as a function of different consolidation conditions.](image)

![Fig. 4 SEM micrographs of sintered bulks as a function of MPC pressure and precompaction pressure showing cracks formed from the corners of the hardness indenters; (a) Sapphire, (b) 0.5 GPa MPC (c) 1.25 GPa MPC (d) 110 MPa uniaxial precompaction + 1.25 GPa MPC, (e) 220 MPa uniaxial precompaction + 1.25 GPa M (f) 330 MPa uniaxial precompaction + 1.25 GPa MPC.](image)
crack and the fine particles resulted in the crack deflection. Microscopic analysis of the MPCed specimens demonstrated that the direction of the crack propagation changed whenever it meets the fine particles in the matrix and propagated around them. Finally, it is expected that the smaller grains and their homogeneous distribution are more prone to the crack propagation. Under a load of 19.6 N, many large cracks caused by an indentation were observed in the MPCed and sintered bulks without precompaction, whereas a few small cracks were present in the bulk MPCed with precompaction and their size was about 15 μm.

The crack length along the four angles may be different due to the deviation in the local microstructures and residual stresses along the surface layer. It was found from the micrograph in Fig. 4 that the crack propagates both in the matrix and along the boundary of the particles. When the crack meets a particle during its propagation, it forces debonding at each particle-matrix interface and then deflects along the interface. It is reported that an increase in fracture toughness can be achieved by crack deflection and crack bridging. Therefore, it is observed that the resistance to crack propagation by the particles causes the increase in the fracture toughness. In addition, the crack propagation was slowed with the increasing volume fraction of particles and affected as well by the surface energy, particle shape and size as well as the particle-matrix interface.

It should be mentioned that the high strength and low dielectric constants of ceramics are attractive for application as a microwave window, but the statistical failure of ceramics severely limits their use. Especially, sapphire has been the best material for a microwave window because of its high strength, absorbed powder and good tolerance to radiation damage. A microwave window includes a transparent disk made of glass or ceramic that lets microwaves through and an attachment device made of metal that is used to attach the transparent disk. Specially processed and mounted sapphire windows have been shown to provide a significant improvement in microwave/radio frequency powder transmission capabilities compared to the current technology.

In this study, the Al2O3 bulks were fabricated by the combination of MPC and a sintering process, and the breakdown voltage testing was performed to analyze the dielectric property. Figure 5 illustrates the breakdown voltage of MPCed bulks as a function of the MPC pressure and precompaction conditions. The breakdown voltage of the MPCed specimen increased with the increasing MPC pressure from 0.5 GPa to 1.25 GPa and the value for the MPCed at 1.25 GPa is much higher than that of the uniaxial compaction. In addition, the breakdown voltage of the precompacted specimen also increased with increasing precompaction pressure. The highest breakdown voltage of 53 kV/cm was achieved in the precompacted specimens at 220 MPa and 330 MPa, whereas the breakdown voltage of the commercial Al2O3 was 35.47 kV/cm. This result means that densification of the bulk without any cracks increased the breakdown voltage.

In order to identify the fracture mechanism of failure during the breakdown voltage testing, the fracture surface of the bulks was also studied using high magnification SEM micrographs as shown in Fig. 6. Figure 6(a) displays the fully formed hole and melted grain during testing. The specimen MPCed at 0.5 GPa and 1.2 GPa (Fig. 6(b) and (c)) shows formation of a deep hole and the grain size increased due to the evolution of a high temperature during the breakdown voltage testing. However, the observed fracture surface was basically different for the precompactedspecimen (Fig. 6(d)). The micrograph shows a shallow hole. The hole formed in the bulk was smaller than that of the bulks without precompaction due to the higher density. The microstructure is coarser as a result of the high temperature during the voltage loading. However, it was difficult to observe any severe cracks which were easily observed in the samples without precompaction. A fracture analysis demonstrated that bulks with a lower density are more prone to failure than the higher density ones; the lower the density, the greater is the probability to develop cracks and holes in it, and consequently, the lower the load it can withstand before fracture.

Another reason is that the increased breakdown voltage in the MPCed and precompactedspecimens is related to the grain size of sintered bulks. Though the commercial Al2O3 bulks showed the highest density of about 96%, the breakdown voltage decreased due to the coarse grain size. The intrinsic defects like the F, F+ center in a single crystalline Al2O3 act as heat generation sites during passing the high power wave by a second electron formation. As a result, cracks can be formed at those sites. Alternatively, the grain boundary can absorb the second electron. Therefore, it can prevent the heat generation. In this study, it is expected that the specimen consolidated by the combination of the MPC and sintering process showed higher breakdown voltage values than that of the commercial Al2O3 due to fine microstructure. Finally, these research results suggest the possible fabrication of the Al2O3 bulk showing improved mechanical properties and breakdown voltage due to the ultra-fine microstructure and higher density by the combination of precompaction, MPC, and the sintering process.
4. Conclusion

The application of the magnetic pulsed compaction for consolidation of Al$_2$O$_3$ nanopowders was successful for developing an ultra-fine microstructure. The homogeneously distributed fine microstructure and higher density in the compacted bulks fabricated by the combined application of precompaction, MPC & sintering showed relatively higher mechanical properties and breakdown voltage than the others. The ultra-fine microstructure of the compacted bulk by the MPC process prevents crack propagation both in the matrix and along the boundary of the particles. The highest density of 94.5%, highest hardness of Hv 1600, and highest breakdown voltage of 53 kV/cm were achieved by a combination of the MPC process and precompaction.

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