

Consolidation of Al₂O₃ Nano-ceramic

Powders for High Power Micro-Wave Window

Whung Whoe Kim

Dept. of Nuclear Materials Technology Development, Korea Atomic Energy Research Institute, 150

Dukjin-dong Yuseong-gu Daejeon, 305-353, Korea

1. INTRODUCTION

In material aspect, the sapphire (Al₂O₃) is very attractive material for window in the high power application. However, the fabrication and following process is very difficult and much expensive. In addition, the intrinsic defects like F, F⁺ center in single crystalline Al₂O₃ act as heat generation sites during passing the high power wave by a second electron generation. And as a result cracks can be formed at the sites. Many researchers have been studied to clarify the breakdown mechanisms of high power wave, and to make the optimal designs of window to prevent the breakdown. However, the researches for the window materials in high power wave are much less. It is reported that powder metallurgy is useful process to fabricate consolidated Al₂O₃ bulk and there are some advantage to use the nano Al₂O₃ powders and dynamic compaction for the fabrication of bulk Al₂O₃.

For the powder metallurgy, the full densification process like sintering is very important. When we use nano ceramic powders, we can reduce the sintering temperature from 0.5T_M to 0.2~0.3T_M. However, it is so difficult to pressurize nano powders with high green density more than 70% of theoretical density, and it has usually less than 50% before sintering due to the very high strength, brittleness, and surface area, etc.. We have already developed a special dynamic compaction method using the magnetic pulsed force which can pressurize ceramic nano powders such as Al₂O₃, ZrO₂, SiO₂, etc. with relative density more than 70%. It is very effective method to make nanostructured compacts with high green density. Especially, the consolidation of powder plays an important role in determining the property of a finished component.

The nano-powder that have to be consolidated at high temperature exposure could cause the nanograins to growth resulting in the loss of the superior properties. Unfortunately, however, processing these nanopowders into fully dense, bulk products that retain the original nanoscale grain size has proven to be difficult, owing to a unique combination of problems [1] 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

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14. ABSTRACT In this study nanostructured a-Al₂O₃ ceramics have been fabricated by the combined application of magnetic pulsed compaction (MPC) and spark plasma sintering (SPS), and their density and hardness properties were investigated. The combined application of the MPC and the SPS produced very fine and dense alumina ceramics. The measured grain size was 330nm being slightly higher than the size of the starting powder (200&#61566;300nm), suggesting a very low degree of grain growth during the MPC and SPS processes and 99.7%, and the relative density was 99.7% being very close to the theoretical density (3.99g/cm³).					
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consolidation method have been evaluated, such as hot pressing [2], sintering with microwave radiation [3, 4], etc. However, none of these methods have been very successfully in producing fully dense bulk products that retain nanoscale grain size.

Therefore, the consolidation of nanopowder without grain growth is scientifically and technologically important. The most important aspect in the compaction of nanopowders is how to achieve full density while simultaneously retaining a nanoscale microstructure. Therefore, the process to compact the nanopowder while conserving the nanostructure is strongly required. In our previous work, we proposed to use magnetic pulsed compaction (MPC) for effective consolidation of nano-powder [5-6]. The known data for pressing nano-sized powders by high pulsed pressure give us an opportunity of getting extremely dense compacts with nanostructure and high mechanical properties.

Many researchers have discussed several times throughout the difficulty of compacting nano-size powders into green bodies that can be easily examined without crumbling. Nanosize α -Al₂O₃ powder is also very difficult to compact, and high pressures are typically required to obtain structurally sound green bodies.

Last year, we reported for the possibility of nano-size Al₂O₃ powder using magnetic pulsed compaction and sintering process. However, the results exhibited some problem to obtain the fully density component. In this study in order to fabricate nanostructured α -alumina ceramics that generally require a sintering temperature higher than 1400°C, a combined process consisting of both the compaction and subsequent sintering techniques have been introduced. One is a magnetic pulsed compaction (MPC) capable of reaching a higher relative density of the compacts of nano powders owing to a sufficiently high pressure in a very short duration (a few microseconds), and the other is a spark plasma sintering (SPS) enabling a compact to be sintered with a high density at relatively low temperatures and in a short duration (typically a few minutes) [7]. The structure, density and hardness properties of the Al₂O₃ ceramics fabricated by the combined MPC and SPS processes have been studied with a comparison of those by the conventional compaction and sintering processes.

2. EXPERIMENTAL PROCEDURE

2-1 Consolidation of Al_2O_3 nanopowder

The starting powder for the compaction was $\alpha\text{-Al}_2\text{O}_3$ with 99.8% purity and 200~300nm in particle diameter, which was supplied by Inframat Advanced Materials Company. 1.5 grams of raw Al_2O_3 powder was loaded into a die and punch unit whose outer and inner diameters were 50 and 15 mm, respectively. The Al_2O_3 powder was consolidated with the shape of disc by magnetic pulsed compaction (MPC).



Fig. 1 showing the general view of MPC equipment

Fig. 1 shows the MPC equipment used in this research (pulsed force: up to 1,000KN, compacting pressure: up to 3 GPa, control temperature: up to 500 °C). A graphite paste was used as a lubricant on the die wall and the bottom punch. The pressure of magnetic pulsed compaction (MPC) was 2.1 GPa in the room temperature. In order to improve the density and properties, the starting powder was pre-compacted in a die under 2.1 GPa and then each precompacted sample was SPSed at high temperature. The MPCed bulks were sintered at 1,450 °C for 3 hrs in an air atmosphere. Five different samples were prepared by using a conventional static compaction (MTS), MPC, SPS, pressureless sintering treatment as summarized in Table 1.

Table 1. The preparation conditions of the samples with various compaction and sintering methods.

Sample No.	Compaction	Sintering
1	Static compaction ($P_c=110$ MPa)	PS ($T=1450$ °C, $t_h=2$ hr, $V_h=6.7$ °C /min, in air)
2	MPC ($P_c=2.1$ GPa)	PS ($T=1450$ °C, $t_h=2$ hr, $V_h=6.7$ °C /min, in air)
3	None	SPS ($T=1350$ °C, $t_h=0$ min, $V_h=100$ °C /min, $P_s=50$ MPa, in vacuum)
4	None	SPS ($T=1350$ °C, $t_h=10$ min, $V_h=100$ °C /min, $P_s=50$ MPa, in vacuum)
5	MPC ($P_c=2.1$ GPa)	SPS ($T=1350$ °C, $t_h=10$ min, $V_h=100$ °C /min, $P_s=50$ MPa, in vacuum)

(MPC: magnetic pulsed compaction, PS: pressureless sintering, SPS: spark plasma sintering, P_c : compaction pressure, P_s : sintering pressure, T : temperature, t_h : holding time, V_h : heating rate)

2-2 Microstructure and properties of MPCed and sintered bulk

The MPCed and sintered bulk was polished using diamond paste and thermally etched at a temperature 100 °C. The apparent density of the bulk was measured by the Archimedes method using water and the values averaged. The relative density was calculated assuming a true density of 3.987 g/cm³ for α -Al₂O₃.

Fracture surface of sintered bulks was observed with a scanning electron microscope (SEM). Grain size analysis was performed on the digitized SEM photographs using image analysis. X-ray diffraction (XRD) patterns were obtained at a scanning rate of 4° /min with 2θ range from 10 to 80° using a fully automated diffraction with Cu K α (0.15406 nm) radiation. Microstructure of sintered bodies were examined by scanning electron microscopy (SEM) to investigate fracture model and grain size. Vickers hardness measurements were performed on a Vickers hardness tester using a Vickers indenter with a load of 19.6 N applied for 10 s.

3. RESULTS AND DISCUSSION

Fig. 2(a) shows the typical morphology of the Al_2O_3 nanopowder particles used in this investigation, as observed by FE-SEM. The powders have a size of approximately 50~200 nm, a smooth surface and elliptical shape. Further, several powder particles seem to consist of the large particles appeared to be formed by agglomeration of smaller particles. Fig. 1(b) shows the morphology of Al_2O_3 nano-powder particles observed by TEM. The powder particles have a size range of approximately 100 nm with spherical and elliptical shape. Microstructural investigation at higher magnification reveals that the size of the Al_2O_3 in coarse area varied between 150 and 200 nm.

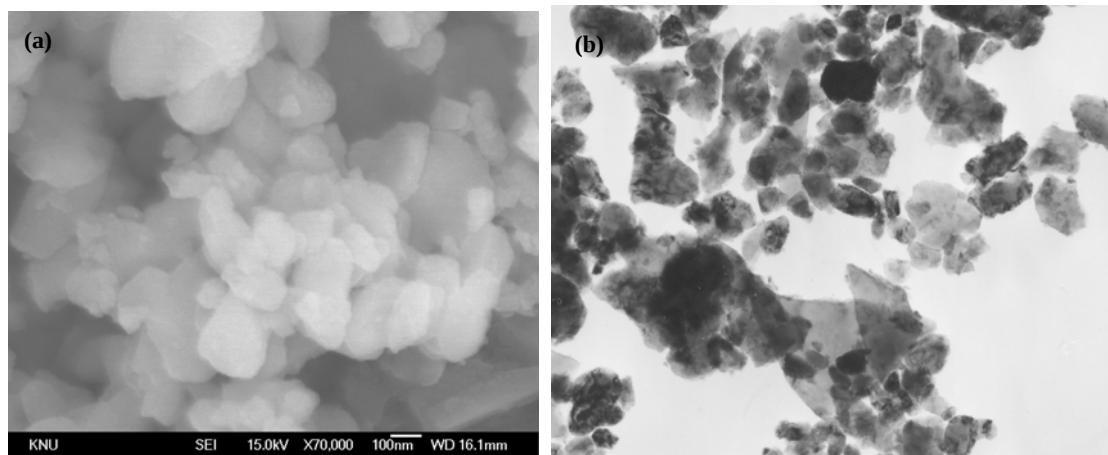


Fig. 2 SEM (a) and TEM (b) morphology of Alumina nanopowder

In order to examine the phase information, nanopowder and bulk are analyzed by XRD. Fig. 3 shows X-ray diffraction patterns of Al_2O_3 nanopowder and bulk, respectively. The as-nanopowder consists of the $\alpha\text{-Al}_2\text{O}_3$ phase. In the case of MPCed and sintered bulk, the XRD peaks became shaper and their peak intensities increased. The peak broadening is attributed to crystallite size refinement in the powder.

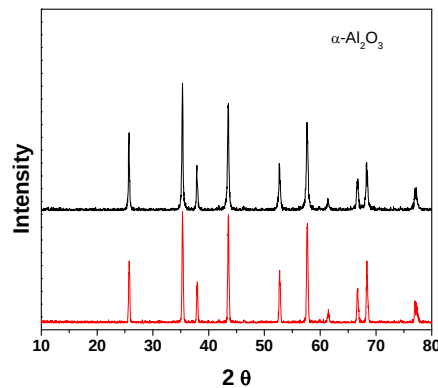


Fig. 3 XRD traces of Alumina nanopowder (up) and bulk (down).

Fig. 4 (a) and (b) show the relative density and Vickers hardness properties, respectively, for the samples prepared by various compaction and sintering conditions as described in Table 1. Samples no. 1 and no. 2 which were prepared by MPC and MTS, respectively, followed by a pressureless sintering exhibited the lowest relative density of about 92%. By the application of the SPS treatment for the alumina powder without introducing any compaction process, the relative density was increased up to about 96~97% (sample no. 3 and no. 4). For sample no. 5 prepared by the combined application of the MPC process followed by SPS, the relative density was increased further, up to about 99.7%, nearly reaching the theoretical density (3.99g/cm^3) of the material. In the sintered ceramics, the degree of densification affects sensitively the final hardness, as clearly seen in Fig. 4 (b). Vickers hardness of sintered bulks shows different values as a function of process. With increasing the density the hardness of sintered bulk was increased. The relative density and the Vickers hardness appear to be correlated well with each other.

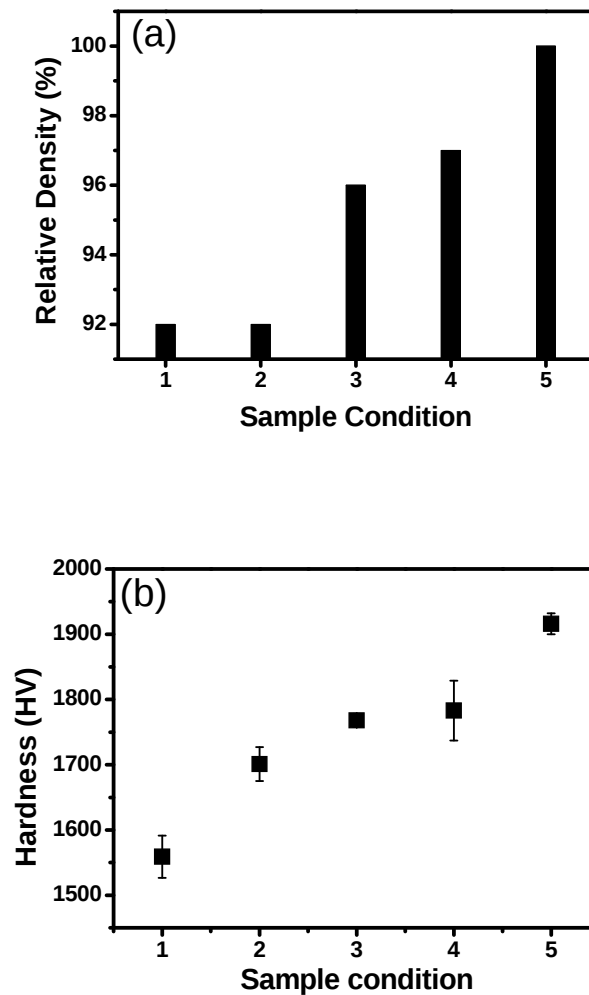


Fig. 4. Variation of (a) the relative density and (b) the hardness for the samples prepared by various compaction and sintering conditions.

It is notable to observe that the density of sample no. 2 by MPC followed by a pressureless sintering was almost similar to that of sample no. 1 by MTS followed by a pressureless sintering, although the hardness of sample no. 2 was higher (more than 140HV) than that of sample no. 1. The different Vickers hardness with process might be associated with the different density distribution of sintered bulks. In addition, the formation of cracking in MPCed specimens can be another important factor leading to different. The relatively low density of sample no. 2 was attributed to the formation of lateral cracks or a peeling at the sintering treatment stage as observed in Fig. 5, probably owing to the excessive strain energy induced by the high compaction pressure of the MPC. In general the densification of the ductile metal particles occurs by a plastic deformation and agglomeration of the particles during the compaction stage. Particularly, a plastic deformation of the particles starts from the contact point between the particles, and as the particle-to-particle contact increases, the compacts become denser with lowering porosity [8]. On the contrary, 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 process could not be readily relaxed, which results in the formation of cracks in the material [9-10]. Therefore, it is necessary to control the strain energy very carefully in the fabrication of ceramic compacts by the application of very high pressure, like the MPC.

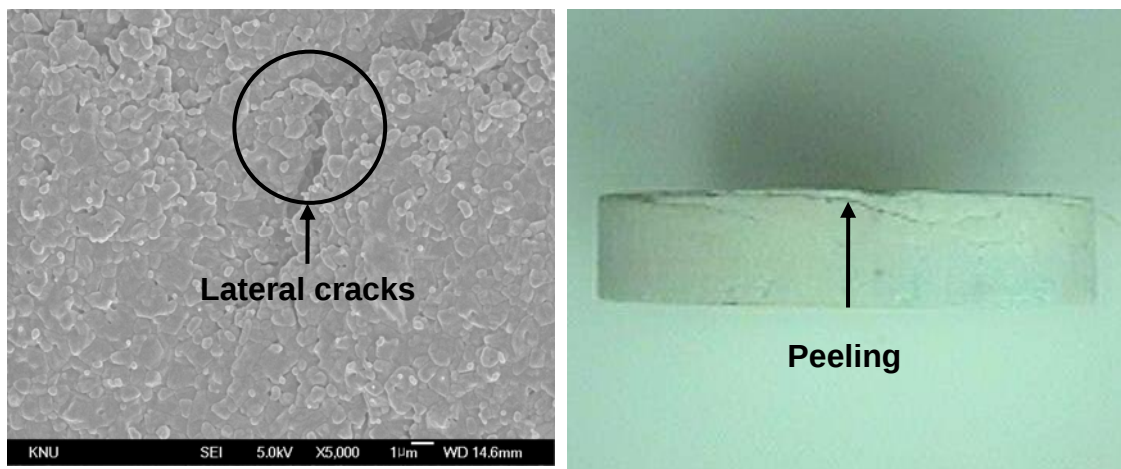


Fig. 5. Formation of the lateral cracks and the induced peeling in sample no. 2 (MPC followed by pressureless sintering).

Fig. 6 (a) to 3 (e) show the FE-SEM images for the fractured surfaced of the five different sintered Al_2O_3 samples. For sample no. 1 prepared by the conventional MTS and a subsequent pressureless sintering, a considerable grain growth was observed with the formation of necks and relatively sharp grain edges (Fig. 6 (a)).

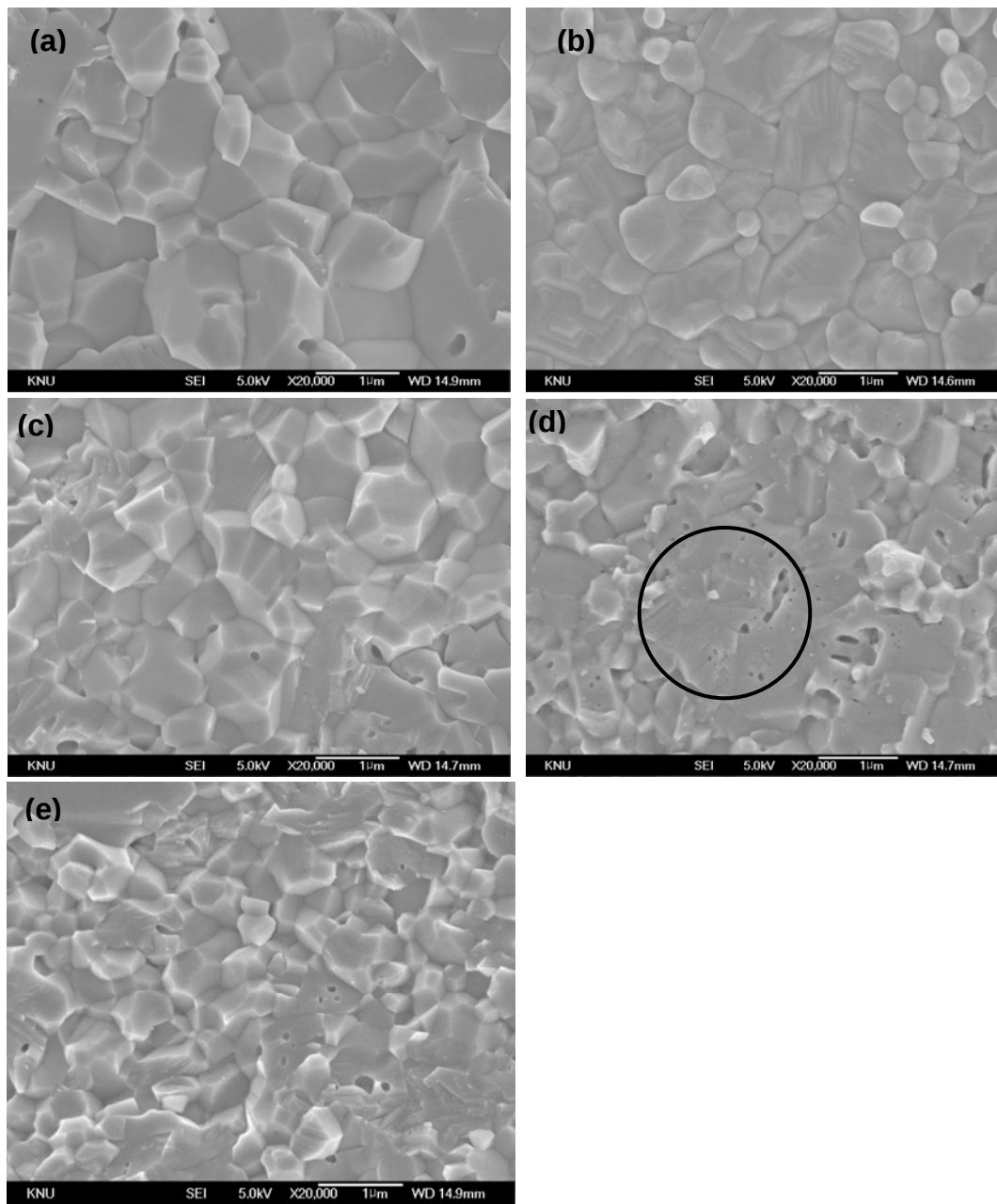


Fig. 6. FE-SEM images of the sintered Al_2O_3 samples with different preparation conditions; (a) MTS + sintering (no. 1), (b) MPC + sintering (no. 2), (c) SPS+ holding 0 min (no. 3), (d) SPS + holding 10 min (no. 4), and (e) MPC + SPS (no. 5).

In sample no. 2 prepared by the MPC followed by a pressureless sintering, however, the grain growth appeared to be less when compared to sample no. 1 and the grains were rounded without forming necks (Fig. 3 (b)), suggesting an insufficient sintering of the particles. Compared to sample no. 1, the smaller grains were formed by the application of the SPS treatment to the alumina powder (sample no. 3 of Fig. 3 (c)). When the SPS holding time at 1350°C was maintained for 10min., a coalescence of the grains was observed from site to site as indicated by the closed circle in Fig, 6

(d). For sample no. 5 prepared by a combined application of the MPC process followed by the SPS, a very fine and dense grain structure was obtained with a sufficient sintering of the particles (Fig. 6 (e)), contributing to a hardness enhancement as already discussed in Fig. 4. For all the samples the average grain sizes were also evaluated from the above FE SEM images by using a computer image analysis and the results are presented in Table 2. It is worthwhile to note that for sample no. 5 prepared by the combined application of the MPC and SPS processes the average grain size was smallest at 330nm, which is almost equivalent to or slightly higher than the size of the starting Al_2O_3 powder. This result suggests that the degree of grain growth was remarkably low during the MPC and SPS processes.

Table 2. Average grain size of the sintered samples by a computer image analysis.

Sample No.	1	2	3	4	5
Average grain size (nm)	~560	~370	~420	~470	~330

Conclusions

In this study nanostructured $\alpha\text{-Al}_2\text{O}_3$ ceramics have been fabricated by the combined application of magnetic pulsed compaction (MPC) and spark plasma sintering (SPS), and their density and hardness properties were investigated. The relative density of the Al_2O_3 by the MPC followed by a pressureless sintering (sample no. 2) was 92 % and almost similar to that of the Al_2O_3 by the MTS followed by a pressureless sintering (sample no. 1), although the hardness of sample no. 2 was higher than that of sample no. 1, due to the formation of lateral cracks or a peeling by an excessive strain energy during the MPC. The combined application of the MPC and the SPS produced very fine and dense alumina ceramics. The measured grain size was 330nm being slightly higher than the size of the starting powder (200~300nm), suggesting a very low degree of grain growth during the MPC and SPS processes and 99.7%, and the relative density was 99.7% being very close to the theoretical density (3.99g/cm^3).

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