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Technical report

Mechanical Properties of Sintered Alumina Ceramics Prepared from Wet Jet Milled Slurries

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Green density of the samples prepared from wet-jet milled slurries was much higher than that of the samples prepared from conventional ball milled slurries indicating that the former method results in closer packing of particles. Due to this close packing, ceramics could be sintered at a lower temperature with little grain growth. Furthermore, ceramics densified to 97% of its theoretical density at 1450°C had a 3-point bending strength of 733 MPa, which is equivalent to that of the Al_2O_3 ceramics prepared by pressure assisted sintering. Wet-jet milling method works well for the dispersion of the particles before molding them into green bodies.

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Key-words : *Slip casting, Milling, Mechanical properties, Strength, Al_2O_3, Structural applications*

1. Introduction

Alumina is widely used as a ceramic for structural applications because of its thermal stability and corrosion resistance;1) however, mechanical properties are still much lower than zirconia and SiAlONs ceramics, to be used reliably in many load bearing applications.

For this purpose, composite materials reinforced by particles2)–7) or fibers8)–10) are preferred. The adverse effect due to the lack of thermal and chemical stability of the secondary phases limited their widespread usage11),12) even though the mechanical requirements are fulfilled. A well known fact is that, mechanical strength of a material depends on the size of the defects present so any decrease in the flaw size would increase strength of the materials.13)

Pressure assisted sintering, such as hot pressing (HP),14) hot isostatic pressing (HIP)15) and spark plasma sintering (SPS)16) are frequently employed if not to remove at least to control the size of the flaws. Bending strength of Al_2O_3 ceramics densified by above approaches reached up to 700 to 800 MPa15),16) as compared to 380 MPa for the normal sintered one.1) However, these methods are not practical as well on account of limitation on the shape and size of the parts to be sintered.

Slip casting method is widely employed in various ceramic forming processes for being suitable to fabricate complex-shaped parts and to scale up at low cost. However, not well dispersed ceramic powders usually results in green density inhomogeneities and defects in the cast body which is hard to heel afterwards. Recently, fabrication of high strength ceramics by slip casting goes along with the aim to achieve a near-net shape manufacturing. To prepare high strength ceramics by slip casting, it is necessary to prepare well-dispersed ceramic slurry to minimize any density inhomogeneities and the flaw sizes after molding.

Wet-jet milling is a novel technique for the pulverization of ceramic powders to prepare ceramic slurries.17) It is reported that the maximum solid loading of the slurries was 50 vol% and the obtained slurries were less viscous and thixotropic. The difference in casting behavior of wet-jet milled and conventional ball milled slurries was discussed elsewhere.18) The green densities of samples prepared from wet-jet milled slurries were higher than that of samples prepared from ball milled ones independent of solids loading. It indicates that ceramic powders were well distributed in the slurries by wet-jet milling, and they were closely packed during slip casting. The obtained green compacts, thus, achieved to higher density compared to the ones prepared by a conventional milling and reached to a high relative density of about 67%.18),19)

In this study, microstructure and mechanical properties of Al_2O_3 ceramics prepared from wet-jet milled and ball milled slurries were studied to account for the observed discrepancy.

2. Experimental procedure

30 vol% high-purity α-Al_2O_3 (AKP–20, Sumitomo Chemical, Japan) with an average particle size of 570 nm was premixed with 70 vol% distilled water and 0.144 mass% NH_4 salt of poly (acrylic acid) (Aron A–6114, Toagosei, Japan) using a planetary homogenizer (AR–250, Thinky, Japan) for 10 min. The mixture was pulverized by a wet-jet milling system (PRE03–20–SP, Genus, Japan). The injection pressure into collision unit and the repeating time of the wet-jet milling were 200 MPa and 5 times, respectively. For comparison, the mixtures were also ground by ball milling for 24 h at 60 rpm in a plastic bottle using high-grade Al_2O_3 balls (10 mmϕ). The obtained slurries were degassed using vacuum pump for 10 min and poured into a gypsum mold. The size of the green body was 100 × 100 × 10 mm. After removed from the mold, the cast green compacts were dried at room temperature for 24 h. Then, they were sintered at 1573 . 1873 K for 2 h in air after pretreatment at 1073 K for 2 h. The sintered specimens were cut by a diamond wheel to the dimension of 3 × 4 × 40 mm for 3-point bending strength measurement.

The relative density of the specimens was measured by the Archimedes technique. The 3-point bending strengths of the sintered test pieces were measured with a span length of 30 mm and crosshead speed of 0.5 mm/min (AGS–10kNG, Shimadzu, Japan). Their surfaces were polished with 0.05 μm alumina pastes. The average bending strength was obtained from measurements of 20 samples. The microstructure images of samples were taken by a field emission scanning electron microscope (FE-SEM) (S–4300, Hitachi, Japan).

3. Results and discussion

3.1 Microstructure

Figure 1 shows the relationship between sintering temperature and achieved relative density of Al_2O_3 ceramics prepared from wet-jet milled (WJ) and ball milled slurries (BM). The green densities of the wet-jet milled and ball milled samples

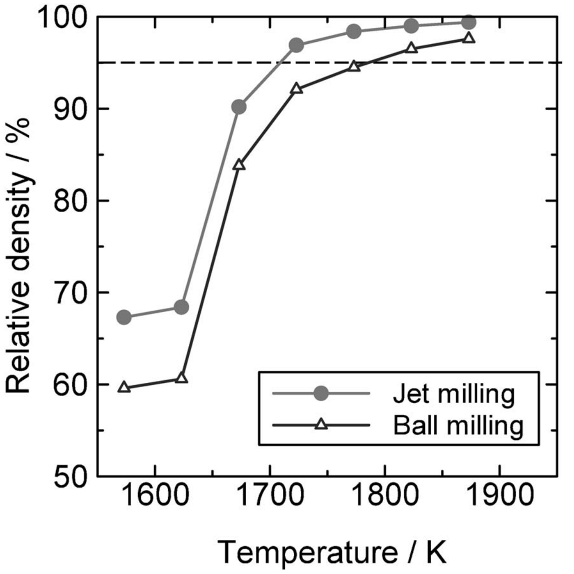

Fig. 1. Relationship between sintering temperature and achieved relative density of Al_2O_3 ceramics prepared from wet-jet milled and ball milled slurries.

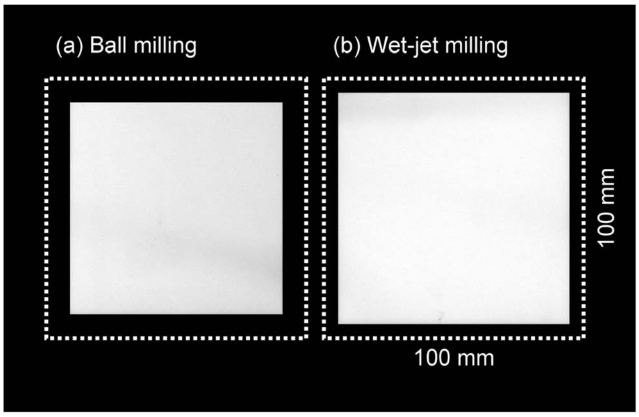

Fig. 2. Microscopic photographs of the (a) ball milled and (b) wet-jet milled samples sintered at 1873 K. The square drawn dotted lines show the original size of the green compacts.

were about 68% and 60%, respectively. Interestingly, samples fired at 1573 K also showed similar values, indicating densities were not affected at all at temperatures below 1573 K. The temperature for the onset of densification was 1623 K. wet-jet milled samples reached to 95% of its theoretical density at a lower temperature (1723 K) than the ball milled ones (1773 K). The difference in density observed at the initial stage of sintering persisted at all sintering temperatures until almost full densification, indicating the importance of dispersion process in ceramic processing.[17] Just from this point, it can be inferred that the wet-jet milling method is, thus, more superior in dispersing ceramic particles than the ball milling method. Therefore, the sintering temperature of the wet-jet milled samples has decreased than that of the ball milled ones because the densification temperature also depends on the density of the green compact.[20]

Figure 2 shows macroscopic image of the compacts sintered at 1873 K (almost full density). The square drawn dotted lines were the original size of green compacts. The linear shrinkage rate of sintered body prepared from wet-jet milled slurries is about 12%, which is much smaller than that prepared from ball-milled ones (about 19%). The difference in the initial green density resulted in with such a difference in sintering behavior.

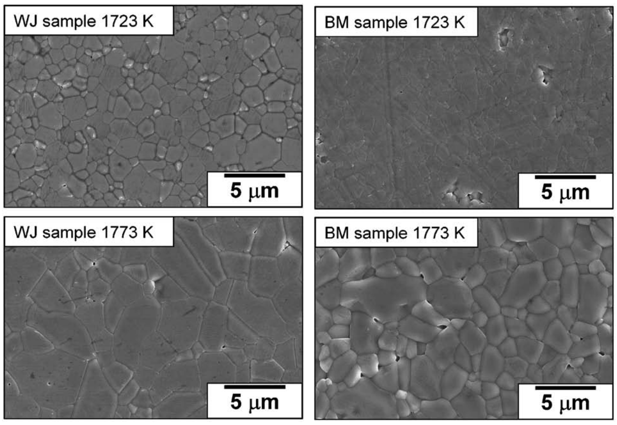

Fig. 3. SEM micrograph of the ball milled (BM) and wet-jet milled (WJ) samples sintered at 1723 and 1773 K.

Table 1. Average Grain Size and Relative Density of the Obtained Al_2O_3 Ceramics

	BM sample		WJ sample	
Sintering temperature [K]	1723	1773	1723	1773
Relative density [%]	92.1	94.5	96.9	98.4
average grain size [μm]	0.7	1.4	1.1	2.8

Figures 3 show the SEM micrographs of the sintered bodies and the average grain sizes estimated from Fig. 3 using the intercept method were listed in **Table 1**. wet-jet milled samples sintered at 1723 K had alumina grains in size range from 0.3 to 3 μm and the average grain size was about 1.1 μm confirming that densification took place with little grain growth. The average grain size increased with increasing sintering temperature as expected, the samples sintered at 1773 K was about 2.8 μm. The key microstructural difference was that the grains were not symmetrical but elongated showing the sign of the abnormal grain growth. Furthermore, the measurement of precise grain size was difficult because of the largeness of the aspect ratio of them. Nevertheless, the presence of grains with more than 50 μm in size was a clear indication of the abnormal grain growth. It is regarded that the suitable sintering temperature for wet-jet milled samples was 1723 K. In the ball milled samples sintered at 1723 K, large pores were observed within the ceramic so, the densification was incomplete. The pores at 1773 K persisted; that is pore size became smaller but did not disappear with the increase of sintering temperature. Even though the pores disappeared by sintering at higher temperatures; some grains grew up into more than 50 μm in size showing abnormal grain growth signs, again. On the other hand, the average grain sizes of ball milled samples were slightly smaller than that of wet-jet milled ones. It is regarded that for particles making many contact points with the others, correlated to the green density, densification and grain growth took place at a relatively lower temperatures compared to the ones not with that many contact points. As a result, the average grain sizes of the wet-jet milled samples were some-

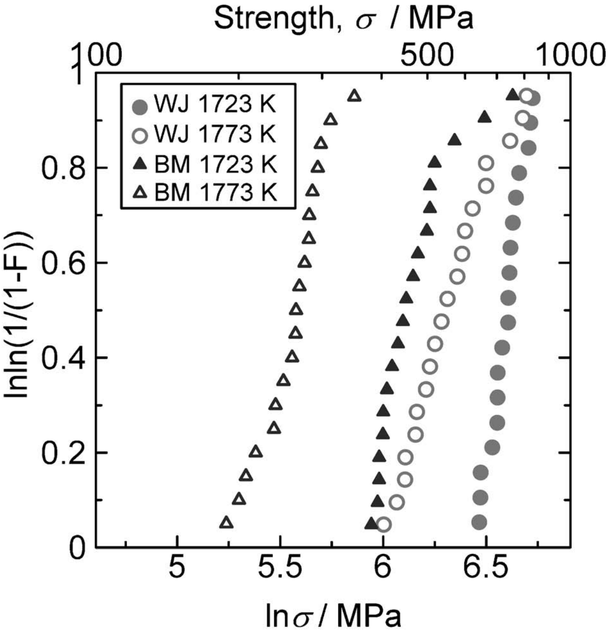

Fig. 4. 3-point bending strength of the ball milled (BM) and wet-jet milled (WJ) samples sintered at 1723 and 1773 K.

what larger than that of ball milled ones.

The sintering process may also be viewed from the point of mass transport between particles in contact with each other either as long distance or short distance. A closely packed particle assembly would definitely easily realize short distance mass transports required for densification at lower temperatures. Similarly, sintering of the ball milled samples doesn't progress at even high temperatures because the contact between particles in ball milled samples were fewer than that of wet-jet milled ones. Thus, heterogeneous microstructure was observed in the ball milled samples with pores.

3.2 Mechanical properties

Figure 4 shows 3-point bending strength of the obtained alumina ceramics. Strength of the ball milled specimens sintered at 1723 and 1773 K was 474 and 262 MPa, respectively, (Weibull module of 7 for both samples) which are fairly lower than that of wet-jet milled samples. This lower strength was caused by a lot of the defects in the samples (Fig. 3). Moreover, because the microstructure was inhomogeneous, the scattering in strength data between individual samples was large. Strength was decreased further due to grain growth though heterogeneous microstructure was improved for the samples sintered at 1773 K.

In contrast, the strength of the wet-jet milled specimens sintered at the optimum sintering temperature of 1723 K was 733 MPa with a high Weibull module of 14. This value is equivalent to Al_2O_3 ceramics prepared by the pressure assisted sintering.15),16)

It was reported that the mechanical strength of ceramics was affected by the $>1\,\mu$m flaws.21) The pores corresponding to these flaws were observed in the ball milled samples sintered at 1723 K (Fig. 3). In contrast, the flaws were not observed in the wet-jet milled ones. It is believed that it is the size of the flaws just mentioned that brought the difference in the mechanical properties of the samples prepared by the two milling methods. The Weibull module of the samples from wet-jet milled slurry is relatively higher than that of the ball milled ones because of the more homogeneous microstructure of the former method. The strength decreased with increasing sintering temperature due to microstructural changes as shown in Fig. 3. Although Al_2O_3 is a low cost material and abundantly available, its application is limited due to the exhibited low mechanical strength. These results confirmed the key features of an ideal microstructure, i.e. small grain size and void free, to be used for mechanical applications. An appropriate processing method has to be employed to achieve these goals. Furthermore, strength of the obtained ceramics strongly depends on the flaw size. These flaws are usually formed during green body formation and survived the sintering process. A well dispersed ceramic slurry and closely packed particles in a homogeneous green body are the key points in preparation of high strength ceramics.

4. Summary

The Al_2O_3 ceramics were prepared from the wet-jet milled slurries. The sintering temperatures of the present samples were 100 K lower than that of the samples prepared by conventional method. The maximum 3-point bending strength of the obtained ceramics was 733 MPa which is very close to the value from samples prepared by pressure assisted sintering methods. It is considered that the decrease in sintering temperatures originated from high green density because of closely packed particles. Wet-jet milling method works well for the dispersion of particles in the slurries for close particle packing during molding process. A better microstructure from wet-jet milled slurries resulted in a homogeneous microstructure after sintering with a high mechanical strength.

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