Microstructural evolution of high purity alumina ceramics prepared by a templated grain growth method

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Textured α-alumina ceramics were prepared by mixing different ratios of three-sized plate-like α-alumina particles and fine equiaxed particles without the use of sintering aids. The plate-like particles have developed a-b planes and were incorporated as an aligned template. Mixed powders were formed into a green sheet using a doctor blade technique and sintered under various conditions (temperature, duration and atmosphere). The development of microstructure and texture in the sintered bodies was examined and correlated with the preparation conditions. The addition of plate-like particles to the fine equiaxed powder suppressed densification and grain growth during sintering. The plate-like particles tend to grow with a lower aspect ratio at high temperatures, especially under vacuum sintering conditions. The addition of 30% plate-like particles produced sintered bodies with the highest uniaxial (001) orientation, but with residual porosity. The addition of 5% plate-like particles resulted in sintered bodies with almost full density but texture development was inferior to that of 30%. The vacuum-sintered specimens with larger amounts of platelets exhibited pseudo-isotropic grain growth and high-to-medium uniaxial (001) orientation, which suggests that anisotropic grain growth is not essential to achieve a high degree of orientation.

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1. Introduction

Single crystal α-alumina is colorless and transparent; however, sintered bodies of α-alumina are typically white in color due to the presence of residual porosity and intergranular phases. The real in-line transmission (RIT) of single crystal α-alumina in the visible light region at a wavelength of 640 nm is 86% at best.1) Light scattering at grain boundaries further reduces the transmission for polycrystalline α-alumina because of its anisotropic refractive index. Therefore, sintered alumina, even without the presence of secondary phases, has no advantage with regard to transparency over optically isotropic materials such as yttrium aluminum garnet (YAG).2) However, translucent polycrystalline α-alumina, which has high mechanical strength and thermal conductivity, is chemically and thermally stable, and is naturally abundant, is considered to be an industrially important material.3)

Since Coble reported the translucency of alumina ceramics prepared with a high-purity raw material in around 1960,4) translucent alumina sintered bodies have been utilized in light-emitting tubes for high-pressure sodium lamps by taking advantage of their mechanical strength and chemical stability. Much effort has been devoted to the development of high-purity and easily sinterable particulate materials, in addition to the development of novel densification processes, such as hot isostatic pressing (HIP)5) and spark plasma sintering (SPS),6) which can reduce residual pores and inhibit grain growth in a sintered body. As a result, the RIT of translucent alumina has been reported to be approximately 70% in recent years.7) Colloidal processing under a strong magnetic field prior to sintering was reported to produce dense alumina ceramics with not only high RIT greater than 50%, but also uniaxial crystal orientation.8,9) Another approach to achieve crystal-oriented ceramics has been the use of anisometric particles.

The fabrication of textured alumina ceramics using flake-like alumina particles as a template was studied by Brandon et al.10,11) and Messing et al.12,13) in the 1990s. Brandon et al. reported grains with uniformly aligned crystal axes, improved flexural strength, and 10% anisotropy of the thermal expansion coefficients by the addition of 5–10% flaky alumina particles with sizes less than 10 μm to granular α-alumina powder.11) Messing et al. examined the addition of approximately 5% CaO and SiO2 along with flaky alumina particles, and concluded that the calcium silicate-based liquid phase acts as a sintering aid at high temperature and is effective for achieving crystal orientation of the sintered body.12) They evaluated the evolution of the grain orientation using X-ray diffraction (XRD) and scanning electron microscopy (SEM) observations and determined that the templates predominantly grow to develop grain orientation, which they expressed as templated grain growth (TGG). Snel et al. then examined the detailed parameters in the doctor blade molding of an aqueous slurry containing a mixture of granular alumina and ca. 10% flaky alumina particles (ca. 3 μm diameter and ca. 0.1 μm thick).13) The shearing force during molding was determined as an important parameter for developing orientation, and the Lotgering factor, which is an index of crystal orientation, was increased to as high as 0.67.

However, systematic experimental studies have not been conducted to observe the development of microstructure and texture in high-purity alumina. The present study is intended to examine the feasibility of the production of dense alumina
ceramics with uniaxially oriented grains by an industrially realizable process. We have selected a sheet forming process using commercially available plate-like alumina particles as templates without the use of sintering aids, which would be applicable to large-sized alumina ceramics for thermal, mechanical and optical applications. In the present study, the effects of the template (size and amount) and the sintering conditions (temperature, duration and atmosphere) on the development of microstructure and texture were examined systematically.

2. Experimental

2.1 Raw materials

Three types of plate-like alumina particles with different sizes (Serath YFA00610, YFA02025, YFA05025, Kinsei Mateco, Ltd.) and high-purity equiaxed-grain α-alumina (Taimicron TM-DAR, Taimei Chemicals Co., Ltd.; BET specific surface area = 13.2 m² g⁻¹, average particle size = 0.17 µm) were used respectively as template powders and matrix powder for the raw material. The average particle diameters and aspect ratios reported by the manufacturer for Serath YFA00610, YFA02025, and YFA05025 are 0.51, 2.11, and 4.62 µm, and 10, 25, 25, respectively. TM-DAR is reported to be sintered up to a density of 3.96 g cm⁻³ by firing at 1350°C for 1 h in air. All the raw materials were subjected to SEM (SU3500, Hitachi High-Technologies) observations and XRD (Ultima IV, Rigaku Corporation, Cu-Kα) measurements to identify the morphology and crystal phases, respectively. Oriented particle monolayer XRD (OPML-XRD) was also conducted for the platelets to determine the developed plane. The specimen names and compositions are listed in Table 1. The names represent the size of plate-like particles and the weight fractions in the specimens, and S, M, and L correspond to YFA00610, YFA02025, and YFA05025, respectively. Control sample was described as Tref which contained only equiaxed-grain α-alumina.

2.2 Forming

The powders were weighed in the ratio in Table 1 for total 20.4 g in each batch and mixed together with 2.2 g of polyvinyl butyral (PVB) and 36.5 ml of organic solvents (ethanol: toluene = 2:3) in a polyethylene pot with alumina balls. After ball-milling for 20 h, the slurry was formed into a sheet with a thickness of ca. 0.5 mm. The sheet was cut at regular intervals and laminated to a thickness of ca. 100 µm. The sheet was embedded in a high purity alumina granulated powder in a silicon carbide (SiC) cylindrical container and sintered at 1700°C for 4 h. The heating and cooling conditions were the same as those used for sintering in air.

2.3 Characterization

The bulk densities of the sintered bodies were measured using the water displacement method. The degree of crystal orientation (F value) for each specimen was evaluated according to the Lotgering method from the XRD diffraction peak intensities for a sintered body that was surface-polished parallel to the original sheet-formed surface. The F values for α-alumina for the (0 0 6) and (1 0 10) crystal planes were calculated using the following equations:

\[ F = \frac{(\rho - \rho_0)}{(1 - \rho_0)} \]

where

\[ \rho = \frac{I}{\Sigma I(h k l)} \]

\[ \rho_0 = \frac{I_0}{\Sigma I_0(h k l)} \]

Here, the subscript in \( \rho_0 \) represents the data for the reference specimen. In the case of (0 0 6), \( I_0 \) is the (0 0 6) peak intensity and \( \Sigma I_0(h k l) \) is the sum of the XRD peak intensities for the reference specimen. Specimen Tref sintered at 1400°C was used as the reference because the XRD pattern had a high intensity and resembled the powder diffraction pattern (PDF01-089-7717). Thirteen main diffraction peaks in the range of 2θ = 10–80° were subject to calculation. Similarly, \( \rho, I \), and \( \Sigma I(h k l) \) represent the values for the test specimens used for evaluation of the degree of orientation.

SEM observations of the plate specimen microstructures were conducted for the fracture surface perpendicular to the original sheet surface. Selected specimens were also mirror-polished and carbon-coated on the section perpendicular to the original sheet surface for evaluation of the crystallographic orientation of individual grains by electron backscatter diffraction (EBSD) analysis.

3. Results

3.1 Characteristics of the raw materials

Figures 1 and 2 show SEM images and XRD patterns for the raw materials, respectively. The hexagonal platelets observed in Figs. 1(b)–1(d) suggest the developed plane corresponds to a-b
plane of the corundum crystal structure. Although the intensities for the (0 0 6) diffraction peak at 2θ = 41.7° are in the order of (d) < (c) < (b), the intensity level was quite low, which suggests the difficulty in aligning these platelets by dry processing. On the other hand, the OPML-XRD pattern for the largest platelets (YFA05025) shows a strong (0 0 6) peak while the other peaks are suppressed in Fig. 2(e), which confirms the developed plane of the platelets is the a-b plane, (0 0 l). However, this tendency for the aligned platelets in the OPML-XRD specimens becomes weaker as the particle size decreases. Except for suppression of the (h k 0) diffractions, the OPML-XRD pattern for the smallest platelets [YFA00610; Fig. 2(g)] is similar to its powder XRD pattern [Fig. 2(d)]. This demonstrates the difficulty in the alignment of small platelets, even by wet processing with a high shear stress.

### 3.2 Density of sintered bodies

Figure 3 shows the relationship between the sintering conditions and the bulk density of the plate specimens sintered in air and under vacuum. The rightmost data of Fig. 3 corresponds to those for the specimens sintered at 1700°C for 20 h in air. Specimen M100 with only medium-sized plate-like particles was difficult to consolidate and contained over 1% open pores, even after sintering at 1700°C for 20 h in air. For the other specimens sintered in air for 4 h, the densities of specimens M60 and M30 reached over 3.9 g·cm⁻³ at 1700°C and open pores were absent. In contrast, the specimens prepared with plate-like particles were low in density when sintered under vacuum, and this tendency was the same for the specimens with a high plate-like particle loading.

The size of the plate-like particles influences the density of the specimen, although there was no significant temperature dependence of the density when sintering above 1400°C in air for the specimens prepared with 5% plate-like particles, which is evident from the results for L5, M5 and S5 shown in Fig. 3. Although the vacuum-sintered L5 and M5 specimens had lower density than the normal-sintered specimens, at 1700°C, S5 reached the same density as Tref by either normal- or vacuum-sintering.

### 3.3 XRD of sintered bodies

Figure 4 shows XRD patterns for the specimens prepared with various amounts of medium-sized plate-like particles by normal sintering. The peak intensity is normalized by the strongest peak. Figures 4(a)–4(d) correspond to the four sintering regimes at 1400–1700°C. Tref showed almost the same XRD profile in any of the firing conditions and it was similar to the reported powder XRD pattern (ICDD: PDF01-089-7717). For the specimens sintered at 1400°C, M30 exhibited the most noticeable change from Tref; the (1 0 10), (0 0 6) and (0 1 8) diffraction peaks became stronger and the other peaks became weaker. This trend became more pronounced for the M30 specimens for higher sintering temperatures and longer times. In the XRD pattern for M5 sintered at 1600°C or above, (1 0 10) was the strongest peak. For the specimens prepared with 5% of plate-like particles, there was no clear relationship between the platelet size and the XRD pattern.

Figure 5 shows XRD patterns for the vacuum-sintered specimens prepared with 5% of different-sized plate-like particles. The peak strength is normalized by the strongest peak. The intensities of the (0 0 6) and (1 0 10) peaks became stronger than that of
Tref, and the (0 0 6) peak intensity was in the order of L5 > M5 > S5, which may reflect the order of ease of platelet alignment.

3.4 Microstructure of sintered body fracture surface

Figures 6 and 7 show SEM images of the fracture surfaces of specimens sintered at 1700°C for 20 h, and four S5 specimens normal-sintered under different conditions, together with Tref, respectively. The vertical direction in each micrograph corresponds to the stacking direction of the formed sheets, so that the length direction of the plate-like particles was expected to align in the horizontal direction and perpendicular to the plane of the sheets.

Figure 6(a) shows the originally 100% plate-like particles coalesced to form large grains with many closed pores trapped inside and no trace of the original plate-like particles. The constituent grains shown in Fig. 6(b) are much smaller than those in Fig. 6(a) and a tendency for long thin grains to align in the horizontal direction is observed. The fracture surfaces shown in Figs. 6(c) and 6(d) are dominated by a transgranular fracture mode, and elongated grains are observed due to the shape of the original plate-like particles. Considering that the medium-sized YFA02025 platelets have an average diameter of ca. 2 μm and an average thickness of ca. 0.08 μm, the grain size in Fig. 6(c) was estimated to be ca. 30 and 5 μm in the horizontal and vertical directions, respectively, which indicates dominant grain growth in the thickness direction. Figures 6(e) and 6(f) comprise five weight percent large and small size plate-like particles. Comparing these photos with Fig. 6(d), the average grain sizes of the sintered bodies were observed to be almost the same but in the order of S5 ≥ M5 ≥ L5 in these normal sintering conditions.
which means drastic grain growth for S5 specimen. Figure 6(g) shows the fracture surface of Tref, which is typical of that observed for a pure alumina sintered body. The size of the equiaxed grains after sintering was over 10 μm, which is approximately 100 times larger than the original TM-DAR particles.

Figures 7(a)–7(d) show the fracture surfaces of S5 sintered under several conditions in air. Figure 7(a) shows the microstructure of S5, which was sintered at 1400°C for 4 h, and is dominated by small equiaxed grains less than 1 μm in size, and no plate-like particles are observed. Figure 7(b) shows significant grain growth up to 10 μm, and grains that are elongated in the horizontal direction are observed. Further elongated grain growth to over 20 μm in length in the horizontal direction and to 5 μm in thickness is observed for the specimen sintered at 1700°C for 4 h [Fig. 7(c)]. Longer heat treatment caused further grain growth [Fig. 7(d)]; however, a quantitative discussion of grain growth is not possible based on the fracture surface. Figure 7(e) shows the microstructure of Tref sintered under the same conditions as S5 in Fig. 7(b), where the same density of 3.96 g·cm⁻³ was obtained. However, the average grain size in Tref was larger than that in S5, which suggests that the incorporation of a small amount of

Fig. 5. XRD profiles for vacuum-sintered specimens (1700°C for 4 h) for (a) L5, (b) M5 and (c) S5.

Fig. 6. SEM images of fracture surfaces for specimens normal-sintered at 1700°C for 20 h. The vertical direction in the micrographs corresponds to the stacking direction of the formed sheets.

Fig. 7. SEM images of fracture surfaces of S5 specimens sintered under four different conditions in air, and of Tref specimen sintered at 1600°C. The vertical direction in the micrographs corresponds to the stacking direction of the formed sheets.
plate-like particles retards the grain growth of alumina. This effect was already observed in a comparison of Tref in Fig. 6(g) with Figs. 6(d)–6(f), where a higher sintering temperature was considered to have promoted not only the exaggerated grain growth of both plate-like particles and fine equiaxed particles but also the retardation of grain growth introduced by the incorporated plate-like particles into the fine equiaxed particles.

Figure 8 shows the fracture surfaces of specimens vacuum-sintered at 1700°C for 4 h, where the vertical direction in the photograph corresponds to the stacking direction of the formed sheets. Figure 8(a) shows a homogeneous porous structure without a trace of the original plate-like particles, with grain growth to more than 5 µm. Figures 8(b) and 8(c) also show equiaxed grain microstructures with similar sized grains as in Fig. 8(a). Necking between the equiaxed grains is more obvious as the amount of plate-like particles is reduced from 100 to 30%. These equiaxed-grain-structured specimens have lower densities of less than 90% with open pores. Figure 8(d) shows a dominantly transgranular fracture mode surface, where in contrast to Figs. 8(a)–8(c), grains elongated in the horizontal direction are observed without open pores. Figure 8(e) shows the elongated grain structure of the vacuum-sintered L5 specimen, where the grain size appears smaller than that in Fig. 8(d), indicating an inverse relationship with the incorporated plate-like particle size. The growth of elongated grains is most remarkable in the S5 specimen, where the grain size was estimated to be ca. 100 µm in length in the horizontal direction and ca. 10 µm thick in the vertical direction. Therefore, the order of the grain size in the sintered specimens prepared with 5% plate-like particles is S5 > M5 > L5, indicating an inverse relationship with the incorporated plate-like particle size. Figure 8(g) shows that the grains in the Tref specimen sintered at 1700°C for 4 h grew to over 30 µm in diameter, which is approximately 200 times larger than the TM-DAR particles.

3.5 Crystal orientation of sintered bodies

Figure 9 shows the relationship between the Lotgering factor $F$ and the sintering conditions for normal-sintered bodies that contain medium-sized plate-like particles. Two Lotgering factors of $F_{(0 \ 0 \ 6)}$ and $F_{(1 \ 0 \ 10)}$ and their summation ($F_{(0 \ 0 \ 6)+\{1 \ 0 \ 10\}}$) are shown as dashed-dotted, dashed and solid lines with the notations C, C1 and C2, respectively. The angle between the (0 0 6) and (1 0 10) crystal planes is 17.5°. Therefore, $F_{(0 \ 0 \ 6)+\{1 \ 0 \ 10\}}$ can be used as an index of the amount of roughly aligned grains with the (00l) planes parallel to the original sheet surface. The maximum $F$ value was attained for M30, and $F_{(0 \ 0 \ 6)+\{1 \ 0 \ 10\}}$ reached 0.78 for the specimen sintered at 1700°C for 20 h.

The relationship between $F$ and the sintering conditions for the specimens prepared with 5% of different-sized plate-like particles is shown in Fig. 10. M5 exhibited higher $F_{(0 \ 0 \ 6)+\{1 \ 0 \ 10\}}$ than both L5 and S5. Figure 10 shows the general tendency that higher sintering temperatures result in higher $F$. In contrast, increased sintering time at 1700°C reduced the $F$ values for the M5 and S5 specimens.

The Lotgering factors for the vacuum-sintered specimens are listed in Table 2. The maximum $F$ value was obtained for M30, and $F_{(0 \ 0 \ 6)+\{1 \ 0 \ 10\}}$ reached 0.30 or above for most of the other specimens. Although three specimens prepared with 5% of plate-like particles had similar $F_{(0 \ 0 \ 6)+\{1 \ 0 \ 10\}}$, the values of $F_{(0 \ 0 \ 6)}$ divided by $F_{(1 \ 0 \ 10)}$ were smaller for smaller plate-like particles, which reflects the difficulty in alignment of small plate-like particles as noted in 3.3.

3.6 Observation of microstructure using EBSD

Figures 11 and 12 show crystal orientation maps obtained from EBSD for specimens sintered in air and vacuum, respectively, at 1700°C for 4 h. The vertical direction in the pattern corresponds to the stacking direction of the formed sheets. Red grains in the
figures indicate that the ⟨001⟩ crystal axis of each grain is oriented within 20° from the vertical direction. These results indicate several features: non-uniformity in terms of both grain shape and crystallographic orientation is present in many of the specimens, the grain size in a vacuum-sintered body is larger than that in the normal-sintered body prepared from the same batch, and the crystal axis orientation of each grain does not necessarily correlate with its shape.

4. Discussion

4.1 Microstructure of normal-sintered ceramics

As shown in Fig. 3, the addition of plate-like particles retards the densification of alumina by normal sintering. Therefore, the use of a small amount of small plate-like particles is desirable to obtain dense sintered bodies of high-purity alumina by the TGG method in the present study. On the other hand, the addition of sintering aids promotes densification, often accompanied by the spontaneous development of plate-like alumina grains in the sintered body.18)

4.2 Texture of normal-sintered ceramics

No evidence of plate-like particles was observed for M100 in Fig. 6(a), which originally consisted of only plate-like particles before sintering. The $F_{(0 0 6)} + F_{(1 0 10)}$ value evaluated from XRD measurements for M100 was ca. 0.2, while that for the same specimen was 0.4 when sintered at 1600°C for 4 h. This indicates that the crystal orientation of the sheet compact remained, even though there was no clear evidence in the microstructure from SEM observations. For M30, which originally contained 30% plate-like particles, the microstructure shown in Fig. 6(c) reveals anisotropic grains that are longer in the horizontal direction, which corresponds well with $F_{(0 0 6)} + F_{(1 0 10)} = 0.78$ shown in Fig. 9. Thus, the fracture surfaces of the M100 and M30 specimens are compared for each sintering temperature in Fig. 13 to discuss the evolution of the grains during sintering.

The grains shown in Fig. 13(a) are ca. 2 μm in length and have an aspect ratio of around 10, corresponding to approximately the same length and half the aspect ratio as those for the original plate-like particles, indicating dominant growth in the thickness direction when sintering at 1400°C. This specimen was not almost densified, i.e., a bulk density was 2.43 g·cm$^{-3}$ and an open

![Fig. 10. Relationship between F value and normal-sintering conditions for specimens prepared with different platelet size particles. The key legend shows the specimen names and Miller indices from which the Lotgering factors were calculated. C, C1 and C2 correspond to $F_{(0 0 6)}$, $F_{(1 0 10)}$ and $F_{(0 0 6) + (1 0 10)}$, respectively.](image)

### Table 2. Lotgering factors for specimens vacuum sintered at 1700°C for 4 h

<table>
<thead>
<tr>
<th>Specimen name</th>
<th>$F_{(0 0 6)}$</th>
<th>$F_{(1 0 10)}$</th>
<th>$F_{(0 0 6)} + F_{(1 0 10)}$</th>
<th>$F_{(0 0 6)} / F_{(1 0 10)}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>M100</td>
<td>0.08</td>
<td>0.34</td>
<td>0.43</td>
<td>0.24</td>
</tr>
<tr>
<td>M60</td>
<td>0.08</td>
<td>0.42</td>
<td>0.50</td>
<td>0.19</td>
</tr>
<tr>
<td>M30</td>
<td>0.12</td>
<td>0.49</td>
<td>0.61</td>
<td>0.24</td>
</tr>
<tr>
<td>M5</td>
<td>0.07</td>
<td>0.23</td>
<td>0.30</td>
<td>0.30</td>
</tr>
<tr>
<td>L5</td>
<td>0.11</td>
<td>0.17</td>
<td>0.28</td>
<td>0.65</td>
</tr>
<tr>
<td>S5</td>
<td>0.04</td>
<td>0.27</td>
<td>0.31</td>
<td>0.15</td>
</tr>
</tbody>
</table>

![Fig. 11. Crystal direction maps obtained using EBSD for specimens sintered in air at 1700°C for 4 h. The vertical direction in the micrographs corresponds to the stacking direction of the formed sheets. Red-colored grains indicate that the ⟨001⟩ crystal axis of each grain is oriented within 20° from the vertical direction in each map.](image)
porosity was 38.5%. The M100 specimen shown in Fig. 13(b) was sintered at 1600°C and has a bulk density of 3.41 g·cm⁻³ and an open porosity of 14.2%, which indicates significant growth in thickness accompanied by growth in the length direction, and tight bonding between grains. This stage is the so-called intermediate stage in sintering theory. The specimen in Fig. 13(c) was sintered at 1700°C and shows further grain growth to form a granular structure with no evidence of the original plate-like particles. The corresponding F_{(0 0 6)/(1 0 10)} values vary from 0.26 for Fig. 13(a), 0.41 for (b) to 0.14 for (c). The sintering mechanism for the pure plate-like particle compact was considered to involve growth in the thickness direction, followed by coalescence of grains to form granular shapes, and further grain growth. The mechanism for the degradation of crystal orientation above 1600°C may be the rearrangement of grains; however, further investigation is required to understand the intermediate stage of sintering.

M30 sintered at 1400°C [Fig. 13(d)] reached a bulk density of 3.44 g·cm⁻³ and an open porosity of 14.3%, which are almost the same as those for M100 sintered at 1600°C [Fig. 13(b)], which indicates the effective role of equiaxed fine particles for densification at relatively low temperatures. Equiaxed particles blended at 70% in the raw material are not distinguishable in Fig. 13(d) and a significant increase in thickness was evident compared with that observed in Fig. 13(a). The specimen in Fig. 13(c), which reached a bulk density of 3.72 g·cm⁻³ and an open porosity of 3.6%, has uniform small elongated grains in the horizontal direction of the micrograph. The specimen shown in Fig. 13(f) was sintered at 1700°C and shows both further anisotropic grain growth and densification. The specimen with an extended sintering time is shown in Fig. 6(c), where a continuous development of the microstructure is observed; however, it is necessary to examine the relationship between the increased orientation and the developed microstructure in more detail.

**4.3 Microstructure of vacuum-sintered ceramics**

As shown in Fig. 3, specimens M100, M60 and M30, which originally contained large amounts of plate-like particles, remained at low density after vacuum sintering compared with the specimens that were sintered at the same temperature in air. However, both the vacuum-sintered specimens with and without the addition of 5% plate-like particles reached densities as high as the normal-sintered specimens. Considering the final stage of sintering, heating in vacuum is advantageous to eliminate closed pores in the sintered body because there is no residual air in the pores. Therefore, densification of the vacuum-sintered specimens with larger amounts of plate-like particles was retarded before the formation of closed pores. The occurrence of neck formation in the vacuum-sintered specimens with ≥30% plate-like particles [Figs. 8(a)–8(c)] suggests that surface diffusion rather than volume diffusion may be dominant under vacuum conditions at 1700°C.

Figures 11 and 12 enable a comparison of the specimen...
microstructures produced by sintering in air and under vacuum, respectively. Whereas the vacuum-sintered M30 in Fig. 12(b) has a porous structure composed of equiaxed grains, the normal-sintered M30 in Fig. 11(b) is dense and composed of small elongated grains. The vacuum-sintered S5 in Fig. 12(e) has a dense structure and significantly large elongated grains are observed, while the normal-sintered S5 in Fig. 11(e) is dense and composed of small anisotropic grains. From this evidence, the vacuum sintering of $\alpha$-alumina is considered to result in significant grain growth compared with normal sintering, which is probably facilitated by the promotion of grain boundary diffusion by the introduction of oxygen defects followed by densification. When significant grain growth occurs before densification, a porous structure with low density would result, as if a coarse powder compact had been used as a starter. For dense specimens such as those shown in Figs. 12(e) and 12(f), significant grain growth may occur by enhanced diffusion.

4.4 Texture of vacuum-sintered ceramics

Figure 14 shows the $F$ values for specimens sintered at 1700°C for 4 h by both normal- and vacuum-sintering. It should be noted that the vacuum-sintered M100, M60, and M30 specimens exhibited relatively high $F$ values, even though their microstructures (Figs. 8 and 12) are dominated by equiaxed grains. This result indicates that anisotropic grain growth is not necessarily an essential condition for macroscopic texture in alumina ceramics.

4.5 Non-uniformity of sintered body microstructure

The layered microstructures shown in Figs. 11 and 12 indicate that there is significant non-uniformity in the original tape-cast sheet, in terms of both the amount and alignment of plate-like particles. For the large and medium-sized platelets (YFA05025 and YFA02025), the weights of the platelets were estimated to be 1000 and 100 times higher than the matrix TM-DAR particles, respectively. Therefore, the sedimentation of heavy platelets may have easily occurred during the sheet forming process.

5. Conclusion

Tailoring techniques for the formation of alumina polycrystalline sintered bodies without the use of sintering aids was investigated. Plate-like corundum particles with a high aspect ratio and developed $a$-$b$ planes were blended with readily sinterable fine equiaxed particles in several ratios, formed into a green sheet by a doctor blade technique, and sintered in air or under vacuum to examine whether and how the sintered body inherits the crystal orientation of the template. Unlike a system with a liquid phase former, plate-like particles tend to grow with a lower aspect ratio at high temperatures. Such pseudo-isotropic grain growth was more notable for the vacuum-sintered specimens with larger amounts of plate-like particles. The addition of plate-like particles to fine equiaxed powder suppresses grain growth and densification during sintering compared to a system with pure equiaxed powder. The degradation of crystal orientation was considered to be attributed to the rearrangement of grains during the intermediate stage of sintering. The addition of 30% plate-like particles produced sintered bodies with the highest texture ($F_{(0006)+(1010)}$ ca. 0.7–0.8), although some pores still remained. The addition of 5% plate-like particles resulted in sintered bodies with full density and medium texture. The texture in ceramics could be improved by optimization of the sheet forming process to achieve uniformly-dispersed and well-aligned plate-like particles in a green sheet.

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References


Fig. 14. $F$ values for specimens sintered at 1700°C for 4 h. Filled and open marks represent normal and vacuum sintering, respectively.