

# Texture Development by Templated Grain Growth in Liquid-Phase-Sintered $\alpha$ -Alumina

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**The distribution and orientation of platelet-shaped particles of  $\alpha$ -alumina in a fine-grained alumina matrix is shown to template texture development via anisotropic grain growth. The textured microstructure ranges from 4 wt% oriented platelet particles in calcined samples to nearly 100% oriented  $\alpha$ - $\text{Al}_2\text{O}_3$  grains after sintering at 1400°C. A  $\text{CaO} + \text{SiO}_2$  liquid phase creates favorable thermodynamic and kinetic conditions for anisotropic grain growth and grain reorientation during sintering. Important criteria for templated grain growth include (1) anisotropic crystal structure and growth, (2) high thermodynamic driving force for template grain growth, and (3) modification of diffusion in the system to continuously provide material to the anisotropically growing template grains.**

## I. Introduction

THE development of textured microstructures in ceramic materials represents a significant departure from the traditional processing goal of dense, fine-grained matrices of equiaxed grains. Traditional microstructures meet the requirements of most applications by averaging the anisotropic properties characteristic of many ceramic crystal structures. In contrast, improved electronic and structural properties could be obtained if polycrystalline ceramics exhibit the anisotropic characteristics typical of single crystals. Such properties can be obtained if the polycrystalline body is textured.

Textured ceramics can be produced by a variety of techniques including sinter forging and eutectic solidification,<sup>1,2</sup> but we propose templated or seeded grain growth as a more general approach. Templated grain growth is defined as the use of aligned particles to obtain texture by grain growth in the anisotropic direction of the template particles. In templated grain growth, large, anisotropic particles are dispersed in a dense, fine-grained matrix. During heat treatment the immediate environment of the template particle favors its growth, as predicted for Ostwald ripening. The sintering environment also satisfies the criteria for exaggerated grain growth set forth by Hillert. The template particles should grow until they impinge on one another or a matrix grain which has coarsened enough to halt the template growth. If the template particles are oriented and grow anisotropically, a textured microstructure should evolve.

Templates can be oriented by various techniques, including tape casting, slip casting, centrifugal casting, and extrusion.<sup>3,4</sup> Recent experiments in  $\text{SiC}$ ,  $\text{Si}_3\text{N}_4$ , mullite, and  $\text{Al}_2\text{O}_3$  have shown oriented template particles initiate textured microstructure development.<sup>5-10</sup>

To develop textured ceramics by templated grain growth, it is important to choose materials which grow anisotropically. Ceramic materials with asymmetric unit cells or a crystal structure consisting of chains or layers of polyhedra often grow anisotropically and have anisotropic surface energies. For example, close-packed atomic planes have low surface energy and are more thermodynamically stable. Such planes also tend to be atomically smooth, making growth perpendicular to the plane difficult to nucleate.

Surface energy anisotropy has been shown to be a sufficient driving force for anisotropic grain growth. Kunaver and Kolar<sup>11</sup> showed that grains with anisotropic surface energies grow anisotropically in a matrix of isotropic grains. Yang *et al.*<sup>12</sup> related the surface energy of grains in an equiaxed matrix to a theoretical anisotropic Wulff plot, and demonstrated that faceted, anisotropic grains develop.

In this paper we have chosen liquid-phase-sintered alumina as an example system to develop an understanding of the principles of templated grain growth.  $\alpha$ -Alumina has a rhombohedral crystal structure of close-packed oxygen planes perpendicular to the *c*-axis with aluminum atoms in 2/3 of the octahedral interstitial sites. Anisotropic growth of  $\alpha$ - $\text{Al}_2\text{O}_3$  is well documented, and is particularly pronounced in the presence of a  $\text{CaO}$ - $\text{SiO}_2$  liquid.<sup>13-19</sup>

Texture development in commercial tape cast alumina substrates was studied extensively in the early 1970s because of the deleterious effect of dielectric constant anisotropy in alumina substrates used in the microelectronics industry.<sup>20-22</sup> Recently, Brandon *et al.*<sup>23-25</sup> oriented  $\alpha$ - $\text{Al}_2\text{O}_3$  platelet particles in an  $\alpha$ - $\text{Al}_2\text{O}_3$  matrix in an effort to reinforce the matrix with the platelet particles. The samples demonstrated improved thermal shock resistance and inhibited crack propagation in the through-thickness direction. Brandon *et al.* attributed texture development to "exaggerated," but equiaxed, growth rather than anisotropic growth, and did not study microstructure evolution. Belmonte *et al.*<sup>26</sup> investigated the sintering behavior of alumina powder compacts containing alumina platelet particles. The  $\alpha$ - $\text{Al}_2\text{O}_3$  platelet particles were shown to inhibit densification, but neither platelet growth nor texture development was measured.

The process detailed in this paper fosters texture development in alumina by combining a templating process with conditions which promote anisotropic grain growth.<sup>27</sup> To obtain dense, fine-grained matrices during heat treatment,  $\alpha$ - $\text{Al}_2\text{O}_3$  seeded, colloidal boehmite sols were selected as the alumina precursor. To promote anisotropic grain growth, calcium aluminosilicate glass formers were added. Hexagonal alumina platelet seed particles served as template particles for growth and were oriented by tape casting. Below, the influence of each component on texture development is examined, and the effect of sintering conditions on microstructure evolution is presented.

## II. Experimental Procedure

Colloidal boehmite sols (15 wt%) were prepared from boehmite powder (Disperal P2, Condea Chemie, GmbH, Hamburg, Germany). Seed particles of  $\alpha$ - $\text{Al}_2\text{O}_3$  were prepared by dispersing high-purity  $\alpha$ - $\text{Al}_2\text{O}_3$  powder (AKP-50 Sumitomo

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Chemical Co. Ltd., Tokyo, Japan) in pH 3 water and centrifuging the dispersion to remove particles larger than  $0.1\text{ }\mu\text{m}$ ; 1.5 wt% of seeds, based on the dry weight of boehmite, were added to provide nucleation sites for the  $\alpha\text{-Al}_2\text{O}_3$  transformation.

CaO and  $\text{SiO}_2$ , high temperature glass formers, were added at a ratio of 1:1, as  $\text{Ca}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$  (Johnson Matthey, Ward Hill, MA) and a colloidal  $\text{SiO}_2$  dispersion (Ludox-AS, Du Pont Co., Wilmington, DE). The  $\text{SiO}_2$ +CaO concentrations were 0.5, 1.0, and 5.0 wt%. Hexagonal  $\alpha\text{-Al}_2\text{O}_3$  platelet particles (Pyrofine Platelets, Elf-Atochem, Pierre-Benite Cedex, France)  $11\text{ }\mu\text{m}$  in diameter and  $1.5\text{ }\mu\text{m}$  thick were added as 4 wt% of the total  $\alpha\text{-Al}_2\text{O}_3$ . Glycerol (7 wt%, Aldrich Chemical Co., Milwaukee, WI) was added as a plasticizer.

Sample processing steps are shown in Fig. 1. After the components were mixed, excess water was removed and the solids + plasticizer concentration was adjusted to approximately 20 vol% by evaporation in a rotary evaporator. The slurries, which had an approximate viscosity of 200 mPa·s at the casting conditions, were then tape cast on a glass plate at a speed of 5 cm/s at a blade height of approximately  $500\text{ }\mu\text{m}$ , then dried for 48 h at ambient conditions. The dry tapes were stripped from the glass, cut and laminated at  $100^\circ\text{C}$ , and 28 MPa for 15 min. Sintering was performed at temperatures between  $1200^\circ$  and  $1600^\circ\text{C}$ , and held at temperature for times between 6 s and 8 h. For SEM analysis, fired samples were polished, and etched with dilute HF to remove the glassy intergranular phase. All samples were inspected by optical microscopy prior to the HF etch, and those fired above  $1400^\circ\text{C}$  appeared to be fully dense. The dimensions and aspect ratios of grains were directly measured from the micrographs (40 measurements for matrix and anisotropic grains) and averaged.

### III. Role of System Components

#### (1) Presence of Liquid Phase

To demonstrate the intrinsic microstructure development of liquid-phase-sintered alumina, a tape cast seeded boehmite sample was sintered at  $1600^\circ\text{C}$  for 6 s. The sample in Fig. 2 is near full density; only the pores entrapped in large grains were present before etching. A number of anisotropic alumina grains are evident, having a wide grain size distribution with an average diameter of  $33\text{ }\mu\text{m}$  and thickness of  $7.9\text{ }\mu\text{m}$ . The average aspect ratio of the anisotropic grains is 4.4. The platelet grains are interspersed in a matrix of less anisotropic grains approximately  $6\text{ }\mu\text{m}$  in diameter. The prevalent faceting and high density of the sample are attributable to the presence of approximately 14.1 vol% liquid at the sintering temperature, as determined from the phase diagram.<sup>28</sup> The heterogeneous microstructure is comparable to those observed by Song and Coble,<sup>16,17</sup> and Kaysser *et al.*,<sup>14</sup> and the faceting is similar to that reported by Simpson and Carter<sup>19</sup> and Handwerker *et al.*<sup>15</sup> in calcia plus silica doped  $\text{Al}_2\text{O}_3$ .

#### (2) Template Grain Growth

A sample containing  $\alpha\text{-Al}_2\text{O}_3$  platelet particles was prepared to demonstrate templating of grain growth. Figure 3 shows a sample which originally contained 10 wt% platelet particles and 5 wt% glass formers. This sample was poured on a glass plate rather than being tape cast, in order to limit the amount of orientation of the platelets. The film was dried to approximately  $500\text{ }\mu\text{m}$ , cut, and fired at  $1500^\circ\text{C}$  for 2 h. Similar to the untemplated case, the liquid phase allowed nearly full densification

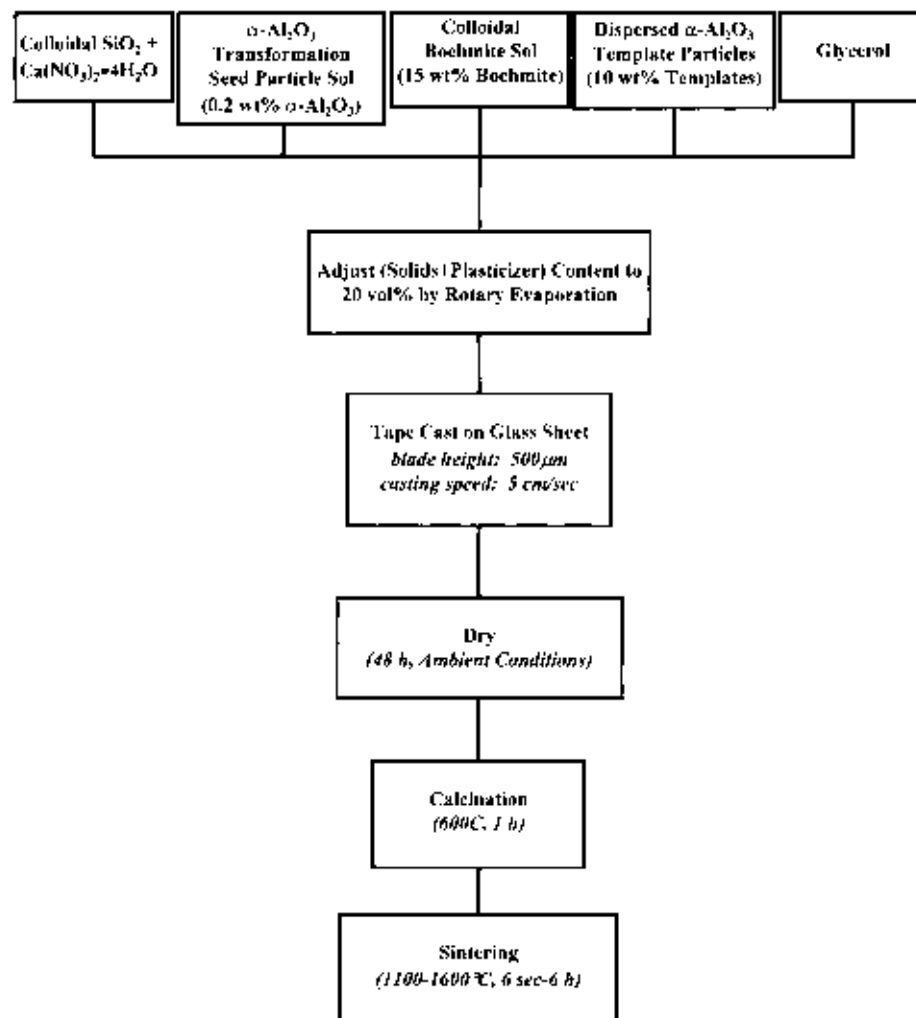
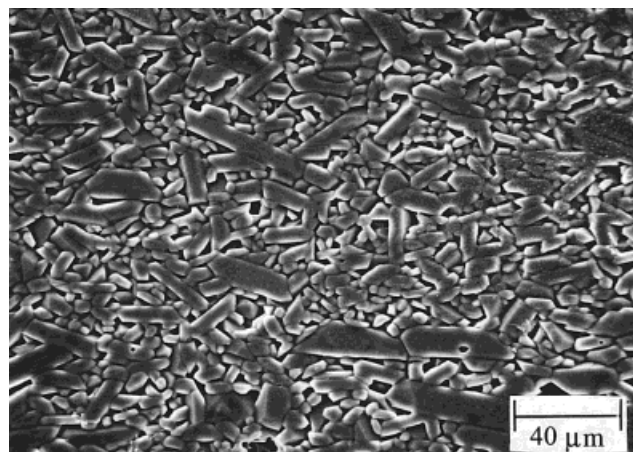


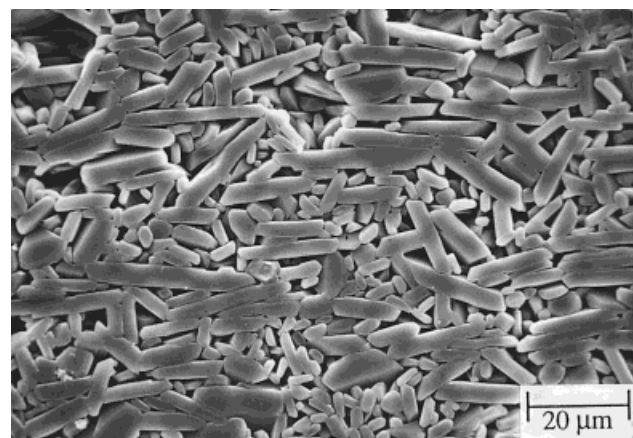
Fig. 1. Processing steps for templated grain growth of alumina.



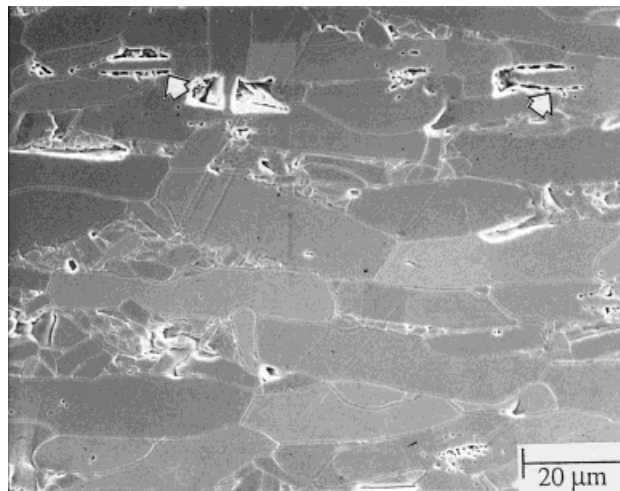
**Fig. 2.** SEM micrograph of untemplated, liquid-phase-sintered alumina at 1600°C, 6 s, 5 wt% CaO + SiO<sub>2</sub>.

and a high degree of grain faceting. The microstructure is characterized by platelet-shaped grains with an average size of 18  $\mu\text{m}$  by 3  $\mu\text{m}$  and an approximate aspect ratio of 6.3. Many template grains settled parallel to the bottom surface resulting in some orientation in the sample. The remaining grains are smaller and less anisotropic, with an average grain size of 4  $\mu\text{m}$ . The grain size distribution and microstructure are similar to those reported by Belmonte *et al.*,<sup>26</sup> and Carisey *et al.*,<sup>24,25</sup> but this sample has much stronger faceting, since it contains a larger amount of liquid phase. The liquid also facilitated extensive anisotropic grain growth. The template particles have become grains with higher aspect ratios compared to the untemplated sample (Fig. 2).

Samples with 10 wt% platelet particles, but no liquid phase formers, were fired at 1600°C for 6 s. The template particles grew anisotropically (Fig. 4), but in contrast with Figs. 2 and 3, none of the grains are faceted. Template growth in Fig. 4 is less regular than earlier figures. Template particles in direct contact with matrix grains grew extensively, but other templates remained isolated from the matrix, because the matrix densified away from the platelet particles, or the template particles constrained densification. Examination of Fig. 4 reveals this difference in the growth at the two ends of single template grains (arrowed). The average length of templated grains in these samples is 35  $\mu\text{m}$ , the average thickness 8  $\mu\text{m}$ , and the average aspect ratio of 4.9. Comparing the microstructure in Fig. 4 with any of the other samples sintered at 1600°C, it is evident that the liquid phase dramatically affects densification, microstructure development, and the faceting of grains.



**Fig. 3.** SEM micrograph of liquid-phase-sintered sample with 10 wt% randomly oriented template particles; 1500°C, 2 h, 5 wt% CaO + SiO<sub>2</sub>.



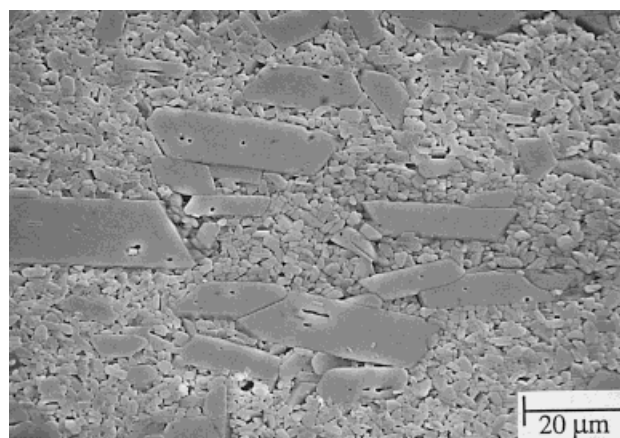
**Fig. 4.** SEM micrograph of sample sintered with 4 wt% oriented template particles; 1600°C, 6 s, 0 wt% CaO + SiO<sub>2</sub>.

### (3) Initial Matrix Grain Size

To further explore the relationship between template particles and matrix grains, samples were prepared with and without  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> seed particles. Nucleation seeds have been shown to control the grain size and density of  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> derived from seeded boehmite sols.<sup>29</sup> In the seeded boehmite case, the matrix grain size is approximately 0.4  $\mu\text{m}$  at 1200°C for the seeding concentration used in this study. For the unseeded boehmite case, samples have fewer nucleation sites, the  $\theta$ - $\alpha$  transformation is delayed, and the resulting grain size is approximately 2–3  $\mu\text{m}$ .

A sample with 5 wt% liquid phase formers, 4 wt%  $\alpha$ -Al<sub>2</sub>O<sub>3</sub> template particles, but no nucleation seeds was fired at 1600°C for 6 s. The dense microstructure in Fig. 5 consists mainly of matrix grains approximately 1.5  $\mu\text{m}$  in size, and unlike the previous samples, only a few well-faceted anisotropic grains developed. The platelet-shaped grains have reached a major axis diameter of 39  $\mu\text{m}$  and a thickness of approximately 13  $\mu\text{m}$  and an aspect ratio of 2.9.

Compared to similar samples with nucleation seeds, the anisotropic grains in this sample have a low aspect ratio. Since the boehmite precursor was not seeded, a coarser Al<sub>2</sub>O<sub>3</sub> matrix developed upon transformation, and grain growth direction was limited by the reduced driving force for solution/reprecipitation. In addition, the increased size of the matrix grains may have also enabled large matrix grains to limit growth



**Fig. 5.** SEM micrograph of liquid-phase-sintered sample with 4 wt% oriented template particles in an unseeded matrix; note the larger matrix grain size; 1600°C, 6 s, 5 wt% CaO + SiO<sub>2</sub>.



of templates through impingement. Similar thickening of anisotropic grains after impingement was reported in  $\text{TiO}_2$ -doped alumina.<sup>30</sup>

#### (4) Orientation Effects

While samples with relatively unoriented templates developed somewhat textured microstructures (Fig. 3), the initial orientation of templates and the final degree of texture development can be markedly improved by tape casting. Figure 6 shows the microstructure of a tape cast sample containing 4 wt% platelet particles and 5 wt% glass formers sintered at 1600°C for 2 h. Compared to previous microstructures, the anisotropic grains are uniformly oriented with hexagonal cross sections. The orientation and growth are striking; the 96% of the microstructure originally composed of equiaxed matrix grains either has been consumed by the initial 4 wt% platelet particles or has adopted the crystal habit and orientation of the template particles.

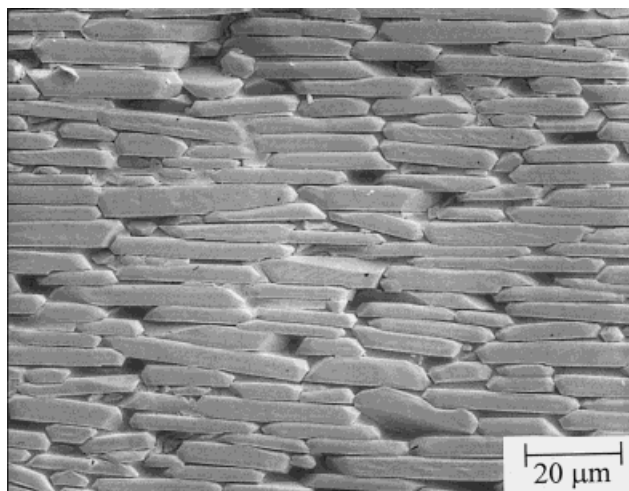
The above studies demonstrate that fine matrix grain size, liquid-phase formers, and oriented template platelet particles dramatically affect the development of highly textured microstructures. Below, we demonstrate how the combination of these conditions controls microstructure evolution and texture development for various sintering regimes.

### IV. Microstructural Texture Development with Changing Sintering Parameters

#### (1) Liquid-Phase Content

The amount of liquid present during sintering can be altered by changing the amount of CaO and  $\text{SiO}_2$ , or the sintering temperature. In this series of samples, the amount of CaO and  $\text{SiO}_2$  was changed, while the CaO: $\text{SiO}_2$  ratio and the sintering conditions remained constant. Samples with 4 wt% template particles and CaO +  $\text{SiO}_2$  contents of 0.5, 1, and 5 wt% were fired at 1650°C for 2 h; the equilibrium liquid-phase contents in the samples at 1650°C were 1.72, 3.44, and 17.2 vol%, respectively.

In Fig. 7(a) the sample completely densified with 1.72 vol% liquid. Anisotropic grains developed from template particles and a textured microstructure resulted. However, some matrix grains and randomly oriented anisotropic grains remain. The anisotropic grains have an average diameter of 15  $\mu\text{m}$  and an average thickness of 4  $\mu\text{m}$ . The average aspect ratio is low, at 4.3. Compared to the sample sintered with no added liquid-phase formers, the anisotropic grains are similar, as they are poorly faceted, irregularly shaped, and have a low aspect ratio. However, there are major differences as well. Grains are smaller



**Fig. 6.** SEM micrograph of liquid-phase-sintered sample with 4 wt% oriented template particles; 1600°C, 2 h, 5 wt% CaO +  $\text{SiO}_2$ .

in both directions, and it is not easy to distinguish grains which formed from matrix grains and those which formed from templates by shape or size. Additionally, the microstructure of Fig. 7(a) does not show packing and densification inhomogeneities found in Fig. 4.

In Fig. 7(b), the sample densified in the presence of 3.44 vol% glass formers developed a fundamentally different microstructure. All grains are anisotropic and faceted; with an average aspect ratio of 6.7. The increased liquid content allowed grains to grow to an average diameter of 21  $\mu\text{m}$  and thickness of 3  $\mu\text{m}$ . However, while many of the grains are aligned in the tape casting direction, some remain unaligned and larger glassy phase pockets reside in the areas of misalignment.

In Fig. 7(c), the sample was densified in the presence of 17.2 vol% liquid. The grains became highly faceted and the microstructure became extremely well aligned during sintering. The average grain diameter remained nearly identical to Fig. 7(b) with an average diameter of 21  $\mu\text{m}$ , but the grains are thicker at approximately 5  $\mu\text{m}$ , resulting in a lower aspect ratio of 4.1. Where grains contact, no grain boundaries are visible. Interestingly, few of the highly faceted anisotropic grains are out of alignment.

The apparent increase in alignment suggests sufficient means for grain rearrangement exist during and after anisotropic growth. The driving force for such rearrangement could be van der Waals or DLVO interactions in the high temperature melt.<sup>31,32</sup> Alternatively, in systems with high liquid contents, misaligned grains are more likely to impinge early in their growth process, resulting in smaller grain sizes. These smaller, misaligned grains become a source of material for larger, aligned template particles.

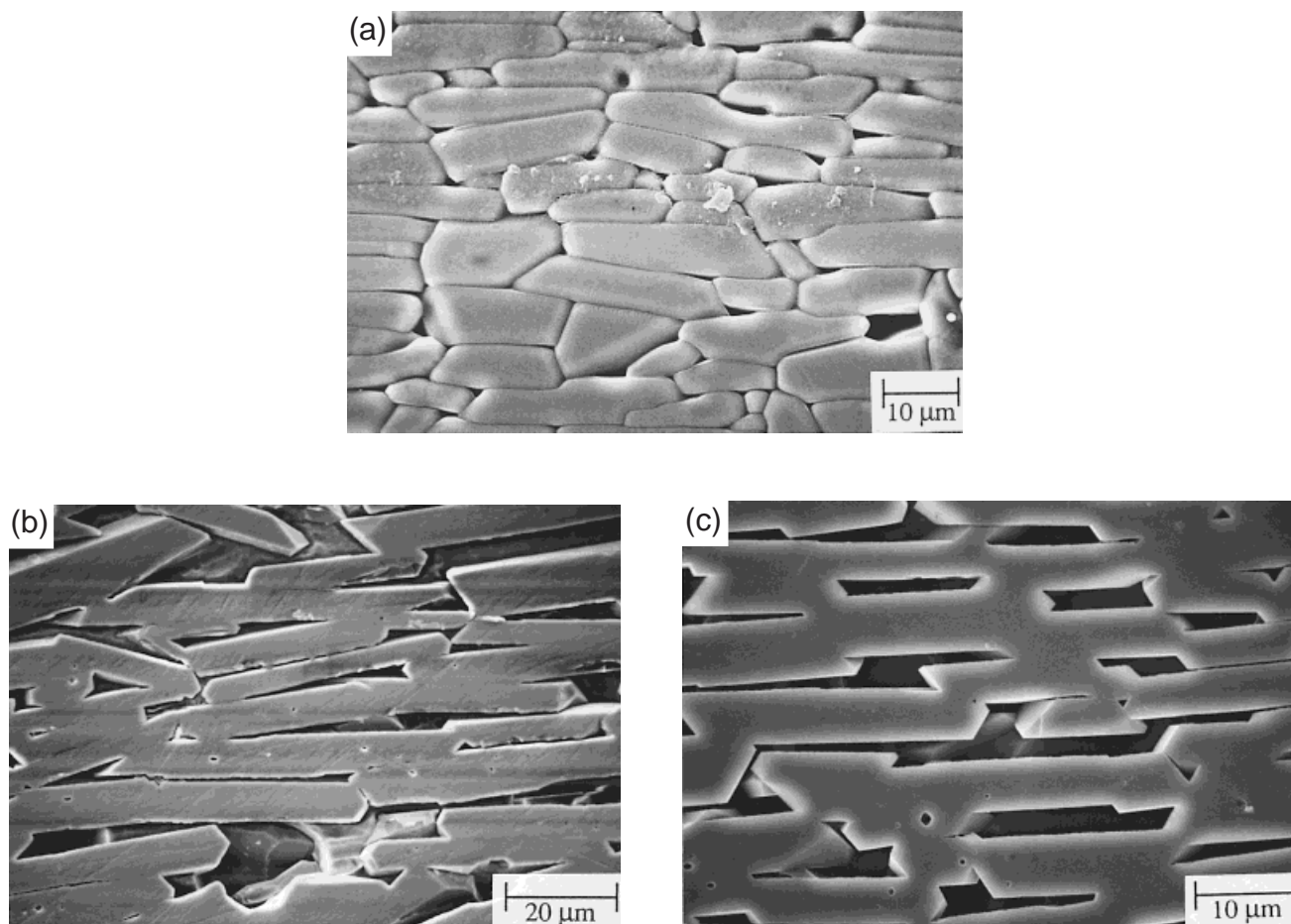
#### (2) Texture Development with Sintering Temperature

Figures 8(a) through (d) show microstructures of four equivalent samples (4 wt% platelet particles, 5 wt% glass formers), sintered at four temperatures (1100°, 1350°, 1400°, and 1650°C) for 2 h. These samples demonstrate how the microstructure develops and grains grow during sintering at different temperatures. In Fig. 8(a) the sintering temperature is 1100°C, which is below the  $\alpha\text{-Al}_2\text{O}_3$  transformation temperature and the eutectic temperature of the glass phase forming system. The template particles are seen to be well aligned by the tape casting process. Platelets with an initial average size of 11  $\mu\text{m}$  by 1  $\mu\text{m}$  and an average aspect ratio of 9 are distributed in an extremely fine matrix of transition alumina, as confirmed by XRD. The only macroscopic porosity is associated with aggregated platelet particles that remain after dispersion and tape casting.

When samples are sintered at 1350°C for 2 h, two phase transformations occur; at  $T > 1200^\circ\text{C}$ , matrix grains transform to  $\alpha\text{-Al}_2\text{O}_3$  and at 1265°C the liquid-phase formers melt to create 10.75 vol% liquid. Compared to Fig. 8(a), both the matrix and template grains in Fig. 8(b) are larger. The platelet particles have grown to an average diameter of 31  $\mu\text{m}$  and a thickness of 3  $\mu\text{m}$  while retaining an average aspect ratio near 9. Porosity is still associated with template aggregates. Basal surfaces remain prominent on the template grains, but the ends of the template grains appear rough. The template grains are well-oriented, but some misorientation present in the initial microstructure is still evident.

Samples sintered at 1400°C develop 11.9 vol% liquid. With the change in temperature and liquid content, major microstructural differences evolve. Matrix grains are completely consumed by an extensive network of anisotropic grains. The sizes and aspect ratios of these grains are difficult to determine because of the interlocking nature of the microstructure. The orientation of the grains appears higher than the 1350°C sample, but several misaligned grains remain. While the sample is dense, some porosity remains concentrated in areas of initial platelet particle misalignment.

In samples sintered at 1650°C for 2 h, 17.2 vol% liquid phase develops and the resulting microstructure which develops differs from the previous samples (Fig. 8(d)). The microstructure consists of extremely well oriented and faceted grains. The



**Fig. 7.** SEM micrographs of liquid-phase-sintered samples with 4 wt% oriented template particles. All samples sintered at 1650°C, 2 h: (a) 0.5 wt% CaO + SiO<sub>2</sub>, (b) 1 wt% CaO + SiO<sub>2</sub>, (c) 5 wt% CaO + SiO<sub>2</sub>.

grains have an average diameter of 21  $\mu\text{m}$  and an average thickness of 5  $\mu\text{m}$ . The aspect ratio of 4.6 for grains in this micrograph is about half of that of grains earlier in the firing process. This change in microstructure may reflect a change in the growth rates of the two crystal directions, or a change in the thermodynamic equilibrium shape of the crystal at this higher temperature.

### (3) Texture Development with Sintering Time

The development of texture over time in samples containing 4 wt% template particles and 5 wt% glass formers was also investigated. Samples were heated to 1600°C and held for 6 s and 6 h (Fig. 9). In Fig. 9(a), the fired microstructure consists of highly oriented, faceted grains, with some grains which are slightly out of alignment. In Fig. 9(b), large, thickened grains have developed, and the residual glass has collected in pockets at the ends of the grains. This pair of micrographs reveals two growth trends which occur after the microstructure consists solely of anisotropic grains. First, anisotropic grains continue to grow by thickening. The aspect ratio decreases from 6 in Fig. 9(a) to almost 5 in Fig. 9(b), and the number of grains in the microstructures decreases with sintering time. Second, the grains appear to have rearranged, or off-axis grains have dissolved and reprecipitated on better aligned grains in the later sample, as the grains in Fig. 9(b) appear to align with one another better.

## V. Criteria for Templated Anisotropic Grain Growth

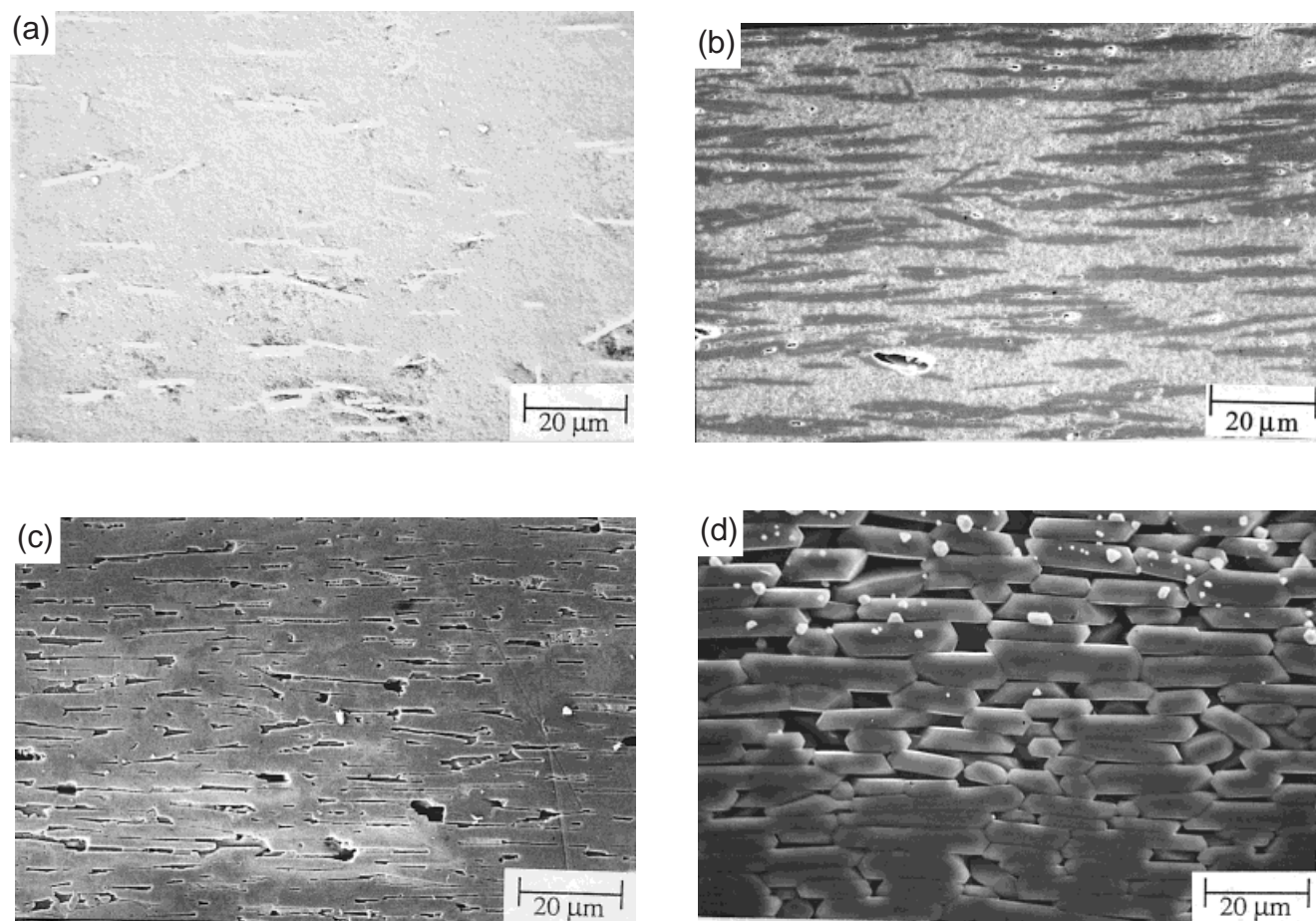
The preceding experiments demonstrated that oriented anisotropic  $\alpha$ -alumina template particles tape cast in a matrix of fine-grained alumina precursors will develop into a highly textured microstructure, especially if the sample is liquid-phase sintered.

Up to this point, no general thermodynamic and kinetic parameters have been proposed. By examining the results of these experiments, it is possible to outline some intrinsic (material properties) and extrinsic (processing parameters) criteria for templated anisotropic grain growth.

The intrinsic criteria for templated anisotropic grain growth are related to the anisotropy of the crystal system in question. For alumina the crystal structure is well known to be anisotropic, and has been characterized to grow anisotropically both in the solid state<sup>33</sup> and in liquid-phase-sintered systems.<sup>14</sup> This tendency to anisotropic growth has also been demonstrated in this work in Figs. 2 and 4. In both figures, the sintered microstructures consist of nonequiaxed platelet grains which have maximized the fraction of the surface area that is basal planes. The slow growth of the basal plane in these and other samples and the prominence of the basal surface in all micrographs attest to the predisposition of the  $\alpha$ -alumina crystal structure to anisotropy under these sintering conditions.

Manipulation of the sintering environment by the presence and orientation of template particles, liquid-phase-former content, and matrix grain size control are all examples of modifying the extrinsic criteria to control anisotropic grain growth. The most obvious extrinsic criteria is the presence and orientation of template particles. By their inclusion, the tendency toward anisotropic grain growth is enhanced. By their orientation, impingement which would normally limit densification in a randomly distributed material and growth is limited. According to Herring,<sup>34</sup> it is unlikely that a large grain will become anisotropic late in its microstructural evolution, because the thermodynamic driving force for achieving an equilibrium anisotropic morphology at large grain size is small. This suggests that anisotropic grains must form early in the densification/growth





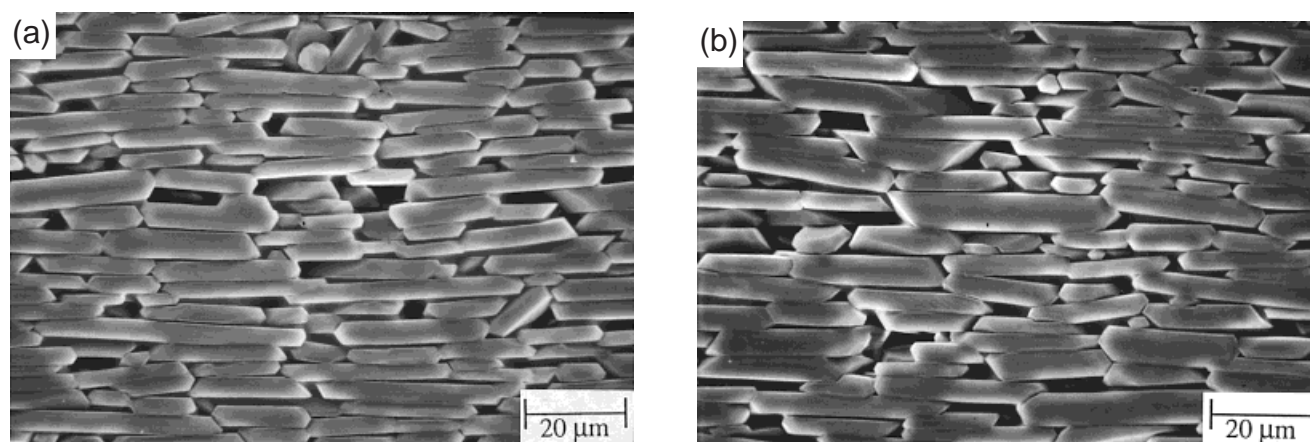
**Fig. 8.** SEM micrographs of liquid-phase-sintered samples with 4 wt% oriented template particles. All samples contain 5 wt% CaO + SiO<sub>2</sub>, and were sintered for 2 h: (a) 1100°, (b) 1350°, (c) 1400°, (d) 1650°C.

process and that large anisotropic particles present in the initial microstructure will preserve their anisotropy, provided material transport to the anisotropic grain is sufficiently high.

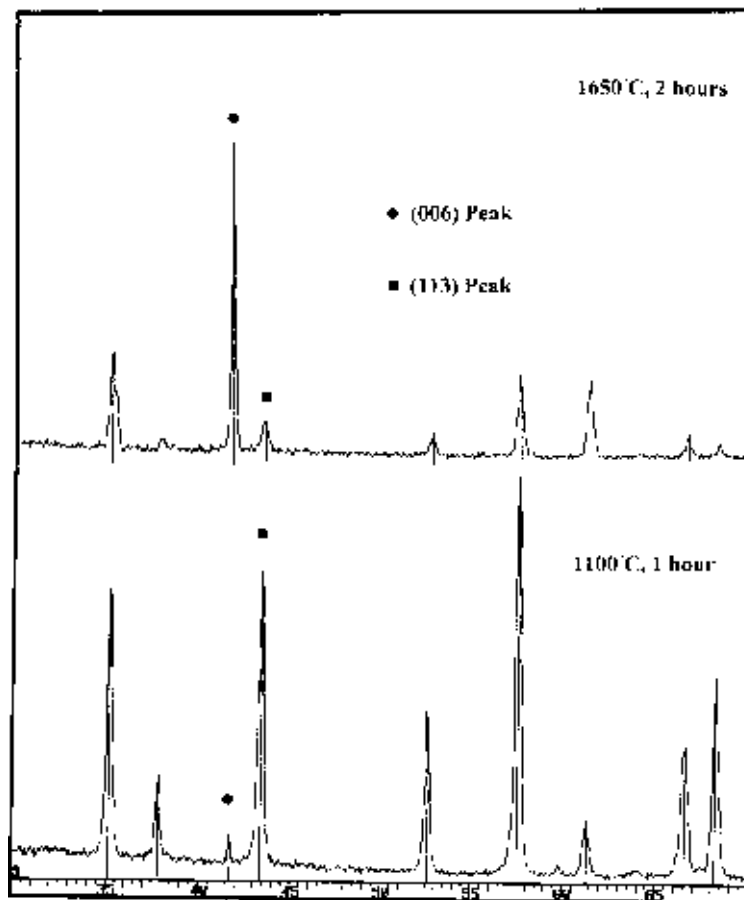
The importance of oriented, anisotropic templates is shown by the difference in aspect ratio between the untemplated microstructure (Fig. 2) and the unoriented template sample (Fig. 3). Both microstructures are strongly faceted, and contain anisotropic grains, but the aspect ratio of the templated microstructure is much higher. In the templated sample, the number of anisotropic grains present was large, and the size of the template grains gave them a size advantage for growth early in

the sintering process. In contrast, in the untemplated sample, a small number of aggregates or anisotropic particles may exist, but even so, have little size advantage over the other grains. Anisotropic growth of such aggregates is easily halted by impingement with an equivalently sized matrix grain. In the templated case, even if impingement occurs almost immediately after growth begins, the template particle will retain a high aspect ratio, due to the relatively slow rate of thickening.

Altering the liquid-phase content was demonstrated to affect the degree of faceting and microstructure development dramatically in Figs. 4, 7, and 8. Determining the exact role of the



**Fig. 9.** SEM micrographs of liquid-phase-sintered samples with 4 wt% oriented template particles; 5 wt% CaO + SiO<sub>2</sub>, 1600°C: (a) 6 s, (b) 2 h, (c) 6 h.



**Fig. 10.** X-ray diffraction patterns for textured alumina samples, fired at 1100°C, 1 h, and 1650°C, 2 h, showing the difference in intensity between (006) and (113) peaks in the two patterns.

liquid phase is a complex problem. Generally, it can be assumed to enhance densification at lower temperatures, given the absence of porosity in liquid-phase-sintered samples before etching. Second, it also acts as a physical homogenizer (as demonstrated by the lack of homogeneity in Fig. 4), allowing material transport to and from all regions of the microstructure. In a liquid-phase-sintered sample, the interfacial energy between the template particle and its environment also chemically homogenizes. The interfacial energy relationship for the templates reduces to a single relationship for each facet and the liquid phase, instead of an assortment of equations for each matrix grain in contact with a facet. Additionally, the liquid phase may also change the growth process of the grains. As increasing liquid phase is present in the system, increased faceting is observed, indicating that a more homogeneous template/liquid phase interface energy relationship exists, as first put forth by Kooy,<sup>35</sup> but also implying that the reactions at this template/liquid interface control the rate of growth in this liquid-phase-sintered system.<sup>36</sup>

Finally, the third extrinsic criteria is the control of thermodynamic driving force through the surface energy of the system. The decrease of interfacial free energy is the driving force for densification, normal grain growth, and anisotropic grain growth. These three processes compete, and if normal grain growth of the matrix continues, anisotropic grain growth will be limited by impingement on large matrix grains. According to Hillert,<sup>37</sup> abnormal grain growth requires a microstructure in which normal grain growth has halted, the average grain size is small, and at least one grain is much larger than the average. On a local scale, these conditions are obtained by the template grains for a sample with a fine-grained matrix (Figs. 6, 7, and 8). However, when the matrix is coarse, the decrease in driving force and increased possibility of template impingement

by a large matrix grain limit longitudinal growth and decrease the aspect ratio, as shown in Fig. 4.

Combined with grain impingement, the decrease in surface free energy from matrix interface area accounts for the gross microstructural difference between highly textured samples that result from fine-grained matrices and the microstructure of isolated anisotropic grains in the coarse-grained matrix (Fig. 5). In the coarse matrix the surface free energy available for growth of the anisotropic grains when the sample is dense is much less than that of systems in which the  $\theta$ - $\alpha$   $\text{Al}_2\text{O}_3$  transformation is seeded.

As a final note on the dependence of texture development in these samples, Fig. 10 shows XRD data for two samples prepared by tape casting  $\alpha$ - $\text{Al}_2\text{O}_3$  powder and platelet particles. Alumina powder was used as the matrix particles for this experiment due to the difficulty in producing a large, flat specimen using boehmite precursors. Fired samples were placed with the top surface of the sample flush with the top of the XRD sample holder.

The lower pattern is from a tape sintered at 1100°C for 1 h. The (006) peak is somewhat higher than a randomly oriented sample, as expected from the oriented platelet particles. However, the largest peak is the (113) peak, as found in a random polycrystalline alumina sample. The upper pattern, from a sample fired at 1650°C for 2 h, shows the radical change in the XRD pattern with anisotropic grain growth. In this pattern, the (006) peak is the largest peak of the pattern, reflecting the fraction of crystals oriented with this face toward the surface of the sample. From this comparison, it is apparent that crystallographic texture development accompanies the growth of the oriented anisotropic grains in these samples.

## VI. Conclusions

Templated grain growth has been shown to develop highly textured ceramic bodies. Preliminary experiments identified the

presence and orientation of template particles, the presence of liquid at the sintering temperature, and the ratio of template to matrix grain size difference as important processing variables. By regulating these processing parameters, control of microstructural texture development becomes a tangible goal.

Large, easily oriented platelets provide templates for the morphology of grains and for texture growth directions. Embedding these platelets in a dense, fine-grained matrix satisfies the Hillert criteria for exaggerated grain growth, since matrix grain growth has stopped, the average grain size is small, and grains much larger than the matrix exist.

Liquid-phase sintering provides kinetic and thermodynamic conditions favorable to anisotropic grain growth. The intragranular liquid phase provides a continuous diffusion path for anisotropic growth and modifies the grain growth kinetics to preserve the anisotropic shape of the template particles. The amount of liquid present during sintering also affects anisotropic grain faceting.

The processing conditions which foster templated anisotropic grain growth have been delineated. Criteria for the anisotropic grain growth, faceting, and texture development have been identified on the basis of the experimental results. Other phenomena which occur during microstructure development, such as apparent grain rearrangement and accommodation with increasing temperature and liquid-phase content, will be more closely examined in future research.

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