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Characterization of Micro- and Macrostructure of Injection-Molded Green Body by Liquid Immersion Method

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Abstract

Micro- and macrostructures of injection-molded alumina body were examined by the liquid immersion method, which is a novel powerful method for characterizing them. Thin specimens for examination were cut from various regions in the green body and made transparent by an immersion liquid. They were then examined by an optical microscope in normal and/or crossed polarized mode. The structural features found by the examination include particle orientation along the flow direction in the injection molding process and few extremely large particles. Their relevance to the forming process and the densification process is discussed. © 1996 Elsevier Science Limited.

1 Introduction

Powder packing structure in a green body is known to have a marked effect on the microstructure development during the subsequent sintering process and is believed to be one of the major factors which govern the properties of the resultant ceramics.[1–5] For establishing the relationship between the forming process and the properties of ceramics, it is necessary to fully understand the powder packing structure of a green body which is formed by a specific method.

This paper presents the particle packing structure of an alumina green body made by the injection molding process. This forming process is known to be very important in mass production of ceramics of complex shape with high accuracy. However, the powder packing structure of the body has not been well understood. Many of the past studies on this molding process have concentrated on the characteristics of the compound, the burn-out process of organics and the properties of ceramics after sintering.[6–9] Orientation of particles is known in a system containing whiskers,[9] but no direct information is available for a more common system consisting of commercial powder with near-equiaxed particle shape.

Features focused on in this paper are the orientation of near-equiaxed matrix particles, and the presence of large particles in very small concentration. We have demonstrated their presence recently in a green body made by a compaction process.[10,11] To study these features, this paper applies the same liquid immersion-polarized light microscopy[11,12] for thin specimens cut from various regions in the injection-molded green body. The specimen is made transparent by the liquid immersion method[5] and is examined with a polarized light microscopy in the transmission mode. Specimens for examination are cut as shown in Fig. 1, since the green body is expected to have a simple structure schematically shown in this figure. Although the major proportion of the powder particles are near-equiaxed, they are considered to have a characteristic shape which is governed by the hexagonal crystal structure of alumina as in large particles. Particles are tentatively assumed to be elongated along a plane normal to the c-axis of the hexagonal system even in small particles of near-equiaxed shape. Grown extensively, the particles should be developed into well-defined platelets. Complete fulfilment of this assumption is not critical in the analysis and discussion of this study, provided a definite relationship is present between the particle shape and the crystal axis.

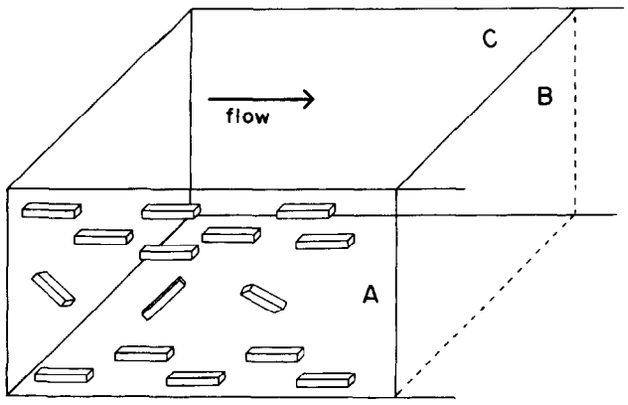

Fig. 1. Structure of green body (schematic) and cutting directions of specimens.

Fig. 2. SEM micrograph of alumina powder particles.

2 Experimental

A commercial compound consisting of alumina particles and acrylic binder was used. The compound was placed in an injection molding machine and made into a green body of a size 100 mm × 50 mm × 5 mm at a nozzle temperature of 150°C by the flow from one end of the body as schematically shown in Fig. 1. After the binder was removed by slow heating (2°C/min) to 450°C, the specimen was heated to 1000°C to provide some strength for subsequent treatment. Thinned specimens for the liquid immersion method (typically 0·2 mm thick) were prepared from various regions of green body as shown in Fig. 1. In the preparation of thin specimens a small piece cut from the green body was polished with sandpaper (#600). After being made transparent by immersion in methylene iodide, the thin specimen was examined with a polarized light microscope in crossed polarized light mode. The green body was also examined by the powder X-ray diffraction analysis and SEM.

3 Results and Discussion

Figure 2 shows a SEM micrograph of powder particles in a green body. The majority of particles constituting the matrix are sub-micron in size and have near-equiaxed shape, but closer examination shows their shape to be slightly elongated. Preferred orientation could not be found in this examination for these small matrix particles. It was also difficult to find large platelet particles which, as will be shown below, were easily detected with the liquid immersion polarized light method. Powder X-ray diffraction analysis showed that the particles are of alpha phase. Essentially the same diffraction patterns were obtained for all specimens of different cut, showing that preferred orientation of particles cannot be detected with this method.

Figure 3 shows the crossed polarized light micrographs taken at two angles of rotation for specimen A cut parallel to the flow direction and perpendicular to the plate surface of the green body. The region near the surface showed a bright–dark change with rotation of the specimen at every 45°, while that near the center remained dark. This result shows that the optical property of the body is uniaxial for the former region and isotropic for the latter region. This optical property is consistent with that expected for the structure shown in Fig. 1.

Large particles were found in both surface and central regions as exemplified in Fig. 3(c) in the observation at high magnification. These scattered particles with exceptionally large size conveniently serve as a indicator showing the direction of orientation for the small matrix particles, for which it is impossible to observe their particle shape or their direction of orientation directly. Those in the former region tended to be aligned with one of their long axes parallel to the flow direction. With the rotation of the specimen, they showed the bright–dark change almost simultaneously with the matrix. Those in the latter region did not show any orientation to a specific direction, nor did they show the bright–dark change at a specific angle with rotation of the specimen. This observation shows that large particles are clearly aligned to the same direction as those of small matrix particles in the surface region. It also shows that both large and small matrix particles are randomly oriented near the central region. These results are again consistent with the structure of Fig. 1.

Figure 4 shows the crossed polarized light micrographs taken at high magnification for the surface and center regions of specimen B cut perpendicular both to the flow direction and the plate surface of the green body. The surface region of the specimen

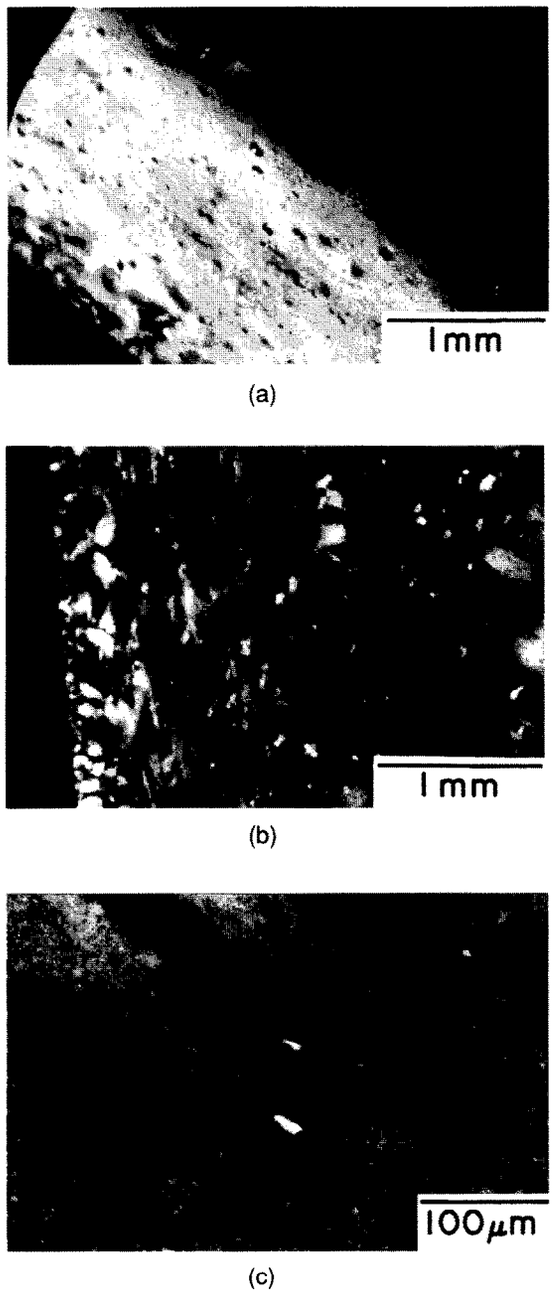

Fig. 3. Injection-molded alumina green body examined by the liquid immersion polarized light microscopy for specimen cut in A plane in Fig. 1. (a) 0°, (b) 45°, (c) detailed structure of (a).

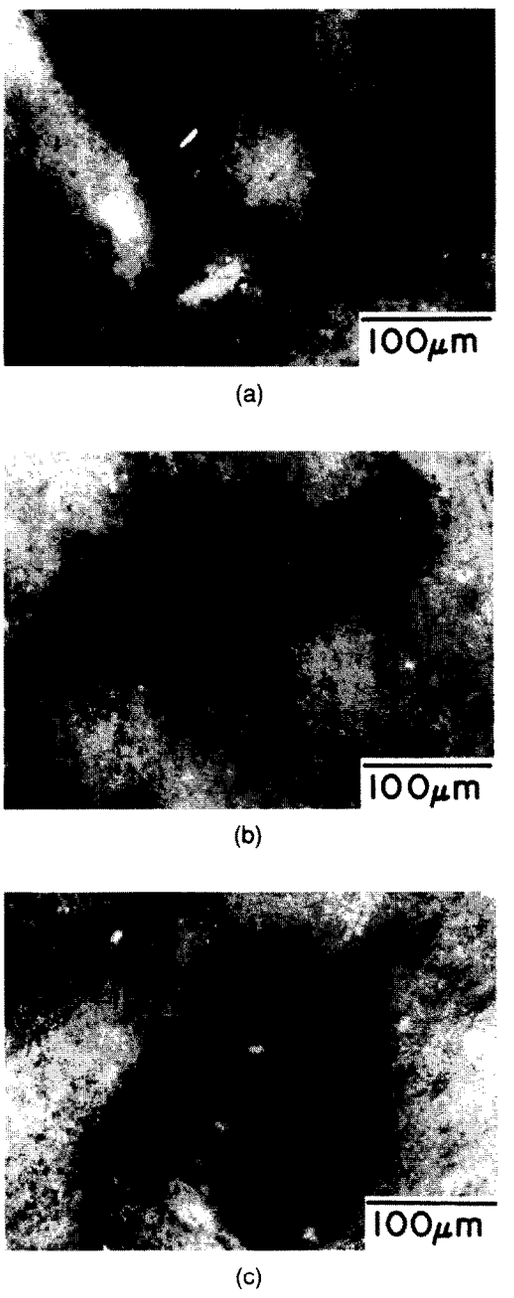

Fig. 4. Injection-molded alumina green body examined by the liquid immersion polarized light microscopy for specimen cut in B plane in Fig. 1. (a) surface region at 0°, (b) surface region at 45°, (c) central region at 0°.

shows the bright–dark change with its rotation and clearly consists of matrix particles aligned along the flow direction. The elongated large particles in the micrograph show that the large alumina plates are aligned with their largest plane parallel to the flow direction revealing their front sides in this observation. The center region of the specimen remained almost dark throughout the rotation. Particles must be oriented randomly in this region. Indeed, some of large alumina particles appear platelet rather than rod-like in this region. This observation again shows that both small and large alumina particles are randomly oriented in this region, supporting the structure shown in Fig. 1.

Figure 5 shows crossed polarized light micrographs taken at two angles of rotation for the specimen C cut parallel to the flow direction near the surface of the green body. Two features are noted in their comparison. One is a large particle at the center of the micrograph (a) and the other is the overall difference in brightness in these micrographs. These results are consistent with the structure shown in Fig. 1. A uniaxial optical property is generated in the green body by the alignment of small matrix particles, causing the

(a)

(b)

Fig. 5. Injection-molded alumina green body examined by the liquid immersion polarized light microscopy for specimen cut in C plane in Fig. 1. (a) 0°, (b) 45°.

bright–dark change at every 45° rotation of the specimen. Large alumina particles are clearly aligned to the same direction as small matrix particles. They show the bright–dark change at approximately the same angle of rotation of the specimen.

All the observations with the polarized light microscope are consistent with the particle packing structure of Fig. 1. The results are in clear contrast to those obtained with conventional analysis including the powder X-ray diffraction technique and SEM microscopy, which failed to reveal the unique structure. The mechanism which produced the structure in the body must be the same as that which formed the particle orientation in a clay body made by extrusion method.[13]

The presence of large alumina particles at very small concentration was again noted in the material used in this study. Their origin can be ascribed to the raw powder used for the experiment. Our experience shows that all alumina powders of industrial grade examined so far contain these large particles. The grinding action appears insufficient to break them into small particles in the conventional procedure adopted for the preparation of the powder-binder compound. They must be eliminated for better processing. Left in the green body, they behave as a potential source of abnormal grain growth in the subsequent sintering process and reduce the quality of the resultant ceramics.

Although the significance of particle orientation on property has not been clarified except for the non-uniform shrinkage in densification and resultant deformation of product, they appear to affect the mechanical property. Local differences of shrinkage in the densification process may create internal strain in the ceramics, reducing their mechanical strength and reliability. Full control of the orientation may contribute to better properties. Marked increase of fracture strength was reported in silicon nitride ceramics with a particle-oriented structure.[14] The significance of particle orientation for mechanical properties is an interesting future research area.

4 Conclusions

The powder packing structure of an injection-molded alumina green body was examined with the novel liquid immersion-polarized light microscopy. Characteristic features found include (1) extremely large alumina particles in very low concentration and (2) particle orientation. The source of the former feature is attributed to large alumina particles in the raw material. They clearly survived the grinding process of the present study and may cause an abnormal grain growth in the final stage of densification. The particle orientation is developed even in a system of nearly equiaxed shape particles by the shear stress in the forming process. It can explain the anisotropic shrinkage and microstructure of ceramics made by the injection molding process. Non-uniform particle orientation, if present, is very unfavorable for ceramics after densification and must be eliminated, since it may cause internal strain and distortion.

References

1. Kingery, W. D., Firing — The proof test for ceramic processing. In *Ceramic Processing Before Firing*, ed. G. Y. Onoda, Jr & L. L. Hench. Wiley, New York, 1978, pp. 291–305.
2. Lange, F. F., Powder processing science and technology for increased reliability. *J. Am. Ceram. Soc.*, **72** (1989) 3–15.

3. Miyashita, M., Kim, J.-Y., Kato, Z., Uchida, N. & Uematsu, K., Effect of compaction pressure on the internal structure of sintered alumina. *J. Ceram. Soc. Japan*, **100** (1992) 1357–1360.
4. Iwamoto, Y., Nomura, H., Sugiura, I., Tsubaki, J., Takahashi, H., Ishikawa, K., Shinohara, N., Okumiya, M., Yamada, T., Kamiya, H. & Uematsu, K., Microstructure evolution and mechanical strength of silicon nitride ceramics. *J. Mater. Res.*, **9** (1994) 1208–1213.
5. Uematsu, K., Miyashita, M., Kim, J.-Y. & Uchida, N., Direct study of the behavior of flaw-forming defect in sintering. *J. Am. Ceram. Soc.*, **75** (1992) 1016–1018.
6. Bandyopadhyay, G. & French, K. W., Effect of powder characteristics on injection molding and burnout cracking. *Am. Ceram. Soc. Bull.*, **73**(3) (1994) 107–114.
7. Zhang, T. & Evans, J. R., Predicting the incidence of voids in the injection molding of thick ceramic sections. *J. Am. Ceram. Soc.*, **76** (1993) 481–486.
8. Pujari, V. K., Effect of powder characteristics on compounding and green microstructure in the injection-molding process. *J. Am. Ceram. Soc.*, **72** (1989) 1981–1984.
9. Tsao, I. & Danforth, S. C., Injection moldable ceramic–ceramic composites: Compacting behavior, whisker degradation and orientation. *Am. Ceram. Soc. Bull.*, **72** (1993) 55–64, 67.
10. Uematsu, K., Tanaka, H., Zhang, Y. & Uchida, N., Liquid immersion–polarized light microscopy as a powerful tool in the research of ceramic processing. *J. Ceram. Soc. Japan*, **101** (1993) 1400–1403.
11. Uematsu, K., Ito, H., Zhang, Y. & Uchida, N., Novel characterization method for the processing of ceramics by polarized light microscope with liquid immersion technique. *Ceramic Trans.*, **54** (1995) 83–89.
12. Uematsu, K., Miyashita, M., Kim, J.-Y., Kato, Zenji & Uchida, N., Effect of forming pressure on the internal structure of alumina green bodies examined with immersion liquid technique. *J. Am. Ceram. Soc.*, **74** (1991) 2170–2174.
13. Robinson, G. C., Extrusion defects. In *Ceramic Processing Before Firing*, ed. G. Y. Onoda, Jr & L. L. Hench. Wiley, New York, 1978. pp. 391–407.
14. Hirao, K., Ohashi, M., Brito, M. E. & Kanzaki, S., Processing strategy for producing highly anisotropic silicon nitride. *J. Am. Ceram. Soc.*, **78** (1995) 1687–1690.