Anisotropic Sintering Shrinkage of Spherical Alumina Particle Compact Aligned in High Magnetic Field

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Spherical alumina particle compacts were prepared by drying alumina slurry in high magnetic field (0–10 T) and cold isostatic pressing (CIP). Anisotropy of shrinkage during sintering was examined for the alumina compacts in detail. The spherical alumina particle compact prepared in 10 T showed sintering shrinkage anisotropy. The sintering shrinkage was larger in the direction parallel to magnetic field direction (i.e., the c-axis direction of alumina crystal) than that in its perpendicular direction. On the other hand, isotropic sintering shrinkage occurred in the compacts prepared in 0 T. The experimental results indicate that the sintering shrinkage of spherical alumina particle compact depends on alumina crystal axis direction. Origin of the sintering shrinkage anisotropy for the spherical alumina particle compacts can be attributed to the particle orientation caused by high magnetic field.

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1. Introduction

Anisotropic shrinkage during sintering was systematically discussed for uniaxially pressed compacts of elongated shape alumina particles in our previous papers. The pressed compacts of elongated shape alumina particles showed the anisotropic shrinkage during sintering, but isotropic shrinkage occurred during sintering in the pressed compacts of spherical alumina particles. Origin of the anisotropic shrinkage was ascribed to particle orientation in the compacts.

However, the effect mechanism of particle orientation on sintering shrinkage has not been understood well. The following two theories may be considered on the sintering shrinkage mechanism of particle orientation at the present time. One is the theory of anisotropic grain boundary number, which was propounded by Coble. Other one is the theory of anisotropic mass transport, which was suggested in our past papers.

In the former theory, the grain boundary number per unit length influences sintering shrinkage, because mass transport to neck occurs along grain boundary during sintering. When alumina particles align with the long axis normal the pressing direction during uniaxial pressing, the grain boundary number per unit length becomes more in the pressing direction, and larger sintering shrinkage occurs in the pressing direction.

In the latter theory, the mass transport rate (or diffusion coefficient) to neck along the grain boundary changes with crystal plane for materials of anisotropic crystal structure, such as alumina, because the atom packing density and surface structure vary with the crystal plane. Alpha alumina has a hexagonal crystal structure of closed-packing oxygen planes (c-plane) perpendicular to c-axis with aluminum atoms in 2/3 of the octahedral interstitial sites. The atom packing is denser in the c-plane than that in a-b plane. The surface energy is lower in the former than that in the latter at temperature above 700°C.

According to the grain boundary number theory, even spherical alumina particle compact aligned should hold isotropic sintering shrinkage, because the grain boundary number is isotropic in the compact. But it should have an anisotropic sintering shrinkage, according to the mass transport rate theory. It is important that examine the sintering shrinkage anisotropy of the spherical alumina particle compact aligned to understand correctly these theories.

It is difficult to align spherical alumina particles with traditional moulding method such as pressing moulding, injection moulding and tape casting etc., which requires elongated shape of particles. In recent years, Sakka et al. developed alignment structure ceramics of spherical alumina particles under high magnetic field, along with application technology development of liquid-helium free superconducting magnet progressing. Alpha alumina crystal has an anisotropic susceptibility, and aligns in high magnetic field with the c-axis parallel to the magnetic field direction.

In this study, spherical alumina particle compacts are prepared by drying alumina slurry in high magnetic field (0–10 T) and cold isostatic pressing (CIP). Anisotropy of shrinkage during sintering is examined for these alumina compacts in detail.

2. Experiments

Spherical alumina particles were used in this study, AA05 (Sumitomo Chemical Company, Japan). It is one of high purity alumina powders (> 99.99% purity), and alpha alumina single crystal with the average particle size 0.5 μm from the manufacturer. The alumina particles and dispersant (0.6 mass%; D305, Chukyouyushi, Japan) were mixed with distilled and deionized water, and ball-milled for 24 h to make alumina slurry (the solid loading: 50 vol%). The slurry was applied to a shallow plate, and dried in a high magnetic field (0–10 T) to make a pellet shape of compact.
the compact was enclosed in a rubber bag, and CIPed at 200 MPa to enhance the homogeneity in particle packing density. The compact was sintered in an electric furnace in air atmosphere at 1600°C for 2–24 h with heating rate 10°C/min. Sintering shrinkage of the compact was determined from its dimension change before and after the sintering in the direction parallel to magnetic field direction (i.e., the c-axis direction of alumina crystal, described as H direction hereafter) and its perpendicular direction (described as D direction hereafter). The green density was measured with a mercury porosimetry (Pore Sizer 9320, Shimadzu). The sintered density was determined with the Archimedes method.

In SEM observation, the plane parallel to magnetic field direction (described as H plane hereafter) and its perpendicular plane (i.e., the c-plane of alumina crystal, described as D plane hereafter) of the sintered body were examined, respectively. The H and D planes of sintered body were finished with diamond slurry (0.5 μm), and thermally etched at 1550°C for 30 min in air atmosphere before the SEM observation.

3. Results

Figures 1 and 2 are the sintered density variations with sintering time at 1600°C for the compacts prepared in 10 T and 0 T, respectively. The green density was 60.0% and 60.8%, for the former and the latter, respectively. The maximum sintered density was 99.4% and 99.7%, for the former and the latter, respectively. The sintered density dropped with increasing sintering time for both the two compacts, when the sintering time exceeded 6 h.

Figure 3 shows the sintering shrinkage change with sintering time in the H and D directions at 1600°C for the compact prepared in 10 T. The sintering shrinkage always was larger in the H direction than that in the D direction at any sintering times. The maximum sintering shrinkage was 16.7% and 15.5%, in the H and D directions, respectively. The compact of particle orientation showed anisotropic sintering shrinkage at any sintering times, although the grain boundary number was same in the two directions.

Figure 4 is the sintering shrinkage change with sintering time in the H and D directions at 1600°C for the compact prepared in 0 T. The sintering shrinkage essentially was same in the two directions at any sintering times. The maximum sintering shrinkage was 15.4%, in both the two directions. Clearly, the compact without particle orientation held isotropic sintering shrinkage.
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Figure 5 is SEM micrographs of the sintered body prepared in 10 T, in the H and D planes at 1600°C for 24 h. There was a clear difference in microstructure in the two planes. In the H plane, grains showed elongated shape, although their initial shape was spherical before sintering. Additionally, the tendency of grain alignment was observed with the short axis (i.e., the c-axis of alumina crystal) parallel to the magnetic field direction. However, shape of grains in the D plane was relatively spherical comparing with that in the H plane.

4. Discussions

Spherical alumina particle compacts were prepared in high magnetic field (0–10 T). The compacts prepared in 10 T showed anisotropic sintering shrinkage, and there was an anisotropic microstructure in their sintered bodies. Particle orientation in the compacts was confirmed with the immersion liquid method of polarized light microscope developed by Uematsu et al. Additionally, grain alignment in the sintered bodies was also observed with X-ray diffraction, the c-plane of alumina crystal corresponded to the D plane in Fig. 5. This was same to Sakka’s result. On the other hand, the compacts prepared in 0 T held isotropic sintering shrinkage, because the particle orientation did not exist in the compacts.

Clearly, sintering shrinkage of spherical alumina particle compact depends on alumina crystal axis direction. The sintering shrinkage is larger in the c-axis direction than that in its perpendicular direction. These results indicate that the mass transport rate to neck is quicker along the c-plane than that along the a-b plane, the diffusion coefficient is larger in the former than that in the latter.

The grain boundary number theory cannot explain the sintering shrinkage anisotropy of spherical alumina particle compact with particle orientation. But it should be emphasized that this study does not exclude importance of the grain boundary number in ceramic compact on the sintering shrinkage anisotropy. In a real system, sintering shrinkage will be affected by crystal axis direction of particle, grain boundary number and particle packing density in ceramic compact. It is difficult to quantitatively analyze relationships among them.

5. Conclusions

Spherical alumina particle compacts were prepared by drying alumina slurry in high magnetic field (0–10 T) and cold isostatic pressing (CIP). Anisotropy of shrinkage during sintering was examined for the alumina compacts in detail. Particle orientation existed in the spherical alumina particle compacts prepared in 10 T, and made them shrink anisotropically during sintering. Sintering shrinkage was larger in the direction parallel to magnetic field direction (i.e., the c-axis direction of alumina crystal) than that in its perpendicular direction. On the other hand, isotropic sintering shrinkage occurred in the spherical alumina particle compacts prepared in 0 T, which did not hold the particle orientation. The experimental results indicate that sintering shrinkage of spherical alumina particle compact depends on alumina crystal axis direction. Origin of the sintering shrinkage anisotropy for the spherical alumina particle compacts can be attributed to the particle orientation caused by high magnetic field.

References