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Effects of heating rate on microstructure and transparency of spark-plasma-sintered alumina

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Abstract

Commercial alumina powder was densified by spark plasma sintering (SPS) at 1150 °C. During SPS processing, the effects of the heating rate were examined on microstructure and transparency. With decreasing heating rate, the grain size and the residual porosity decreased, while the transparency increased. At a heating rate of 2 °C/min, the grain size was 0.29 μm, and the in-line transmission was 46% for a wavelength of 640 nm. The mechanisms for the fine microstructure and low porosity at slow heating, which are conflicting with some existing results, were explained by considering the role of defect concentration and grain-boundary diffusion during densification.

Keywords: Grain growth; Sintering; Porosity; Optical properties; Al_2O_3

1. Introduction

For attaining transparency in fine-grained alumina ceramics, a full density or an extremely low porosity is indispensable. Since residual pores have a significant negative effect on light transmission, for transparent alumina, porosity should generally be reduced to less than 0.05%.[1] Low porosity also allows good mechanical properties such as strength, wear resistance and hardness. In order to achieve such dense and fine microstructures in alumina, hot isostatic pressing (HIP) has widely been used, which effectively eliminates residual pores at low temperatures (1200–1300 °C).[1–4] By using HIP, Krell et al.[1] and Apetz and Bruggen[4] obtained a transparent alumina with an in-line transmission of 50–70%. Their grain sizes are 0.4–0.7 μm and porosities are less than 0.05%. Until quite recently, HIP has been the only way to obtain a transparent alumina with submicrometer grains.

On the other hand, spark plasma sintering (SPS) has recently been paid attention as an alternative method to obtain dense and fine-grained ceramics at low temperatures. Owing to the advantage of rapid heating, the alumina ceramics obtained by SPS have a grain size and density comparable to those of HIPed ones.[5–9] For example, a fully dense (a relative density of ∼100%) alumina with a grain size of 0.5 μm was obtained at 1200 °C by SPS.[8] During SPS of the alumina ceramics, the heating rate was very high (≥150 °C/min) and the holding time at sintering temperature was short (3–10 min). The short heating time at low temperatures significantly suppresses grain growth, and in the case of alumina (a non-conductor), rapid densification can proceed by easy shear-sliding between small powder particles under applied mechanical pressure. By using SPS, Dobedoe et al.[10] obtained a transparent alumina at 1200 °C, although the heating rate and the sintering time were not described in their research paper.

On the other hand, we recently reported that a transparent alumina can be obtained at a low heating rate (2 °C/min), not at a high heating rate (100 °C/min) during SPS.[11] This is the result opposite to the existing researches, although the reason is unclear. For understanding the mechanism, in the present study, we examined the effects of the heating rate on the microstructures and transparency during SPS of alumina. By investigating the microsturctural changes depending on the heating rate, we aimed to understand the phenomena which occur during the sintering process.

2. Experimental procedure

Commercial α-Al_2O_3 powder (TM-DAR, Taimei Chemicals Co. Ltd., Japan), with a purity of 99.99% and an average particle size of 0.15 μm, was used in this study. As-received powder was heated directly, without special treatment or additives, to

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1150 °C under a uniaxial pressure of 80 MPa using a Spark Plasma Sintering machine (SPS-1050, Sumitomo) with a pulse duration of 3.4 ms. Heating was conducted using a sequence consisting of twelve DC pulses (40.8 ms) followed by zero current for 6.8 ms. The heating rate from 600 °C to 1150 °C was varied between 2 °C/min and 100 °C/min. The temperature was measured with an optical pyrometer focused on the non-through hole (1 mm in diameter and 2 mm in depth) of a graphite die. After holding for 20 min at the sintering temperature and subsequent annealing at 1000 °C for 10 min, we obtained a sintered disk with a diameter of 30 mm and a thickness of 3 mm. In addition, for a heating rate of 8 °C/min and 50 °C/min, the sintering time was varied between 0 h and 5 h, in order to examine the grain growth behavior. The mechanical pressure was unloaded before annealing.

The center of the sintered body was machined to a tile of 10 mm × 10 mm with a thickness of 1 mm, and mirror-polished carefully on both sides using diamond slurry. The final thickness of the sample is about 0.9 mm. The in-line transmission was measured in the wavelength range from 0.24 μm to 1.6 μm using a double-beam spectrophotometer (SolidSpec-3700DUV, Shimadzu). The distance between the sample and the detector is about 55 cm.

The microstructure was observed on the specimen surfaces, which had been polished and thermally etched at 1050 °C for 1 h, by using a scanning electron microscope (SEM) (JSM-6500, JEOL). The porosity was measured on the SEM images taken at a magnification of 10,000 times. We did not measure the absolute density because the conventional techniques such as the Archimedes method are insensitive to extremely low porosity. The grain size was measured by obtaining the average cross-section area per grain and assuming spherical grains. The measured grain size is an apparent one, so that it was multiplied by 1.225 to determine the true grain size.[4]

3. Results and discussion

3.1. Grain size

For the alumina sintered at 1150 °C for 20 min, the grain size was smaller at lower heating rates. The dependence of the grain size on the heating rate is shown in Fig. 1, where the grain size decreases with decreasing heating rate. The grain size was 0.55 μm at a heating rate of 100 °C/min, and it was decreased to 0.29 μm at 10 °C/min. Further decrease in the heating rate below 10 °C/min has no remarkable effect on the grain size. At a heating rate of 2 °C/min, a slight tendency toward increasing grain size was observed owing to the increased heating time. The microstructures of the sintered aluminas are shown in Fig. 2.

For the effect of the heating rate on the grain size, there have been conflicting results. Stanciu et al.,[6] Shen et al.[8] and Zhou et al.[9] reported that the grain size of alumina decreased with increasing heating rate, whereas Murayama and Shin[12] reported opposite results that are consistent with the present study. The origin of the conflicting results has been unclear. However, their respective SPS experiments were conducted under different sintering conditions with different Al_2O_3 powders, as shown in Table 1. Although the present Al_2O_3 powder is identical to that of Zhou et al.,[9] the pressure is different, which also has a significant effect on the sintered microstructure. The different sintering conditions may provide a clue for understanding the conflicting results.

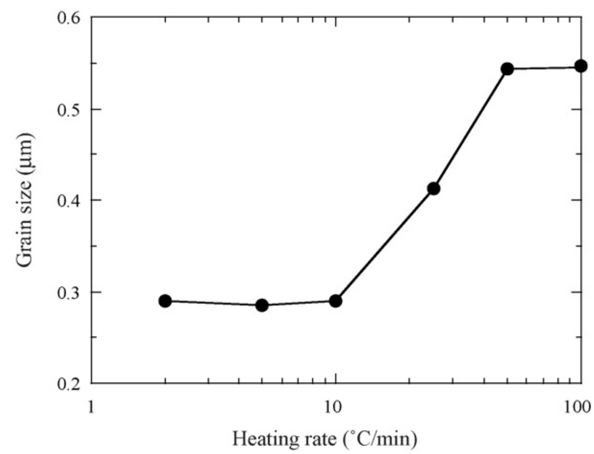

Fig. 1. Dependence of the grain size on the heating rate for sintering at 1150 °C for 20 min.

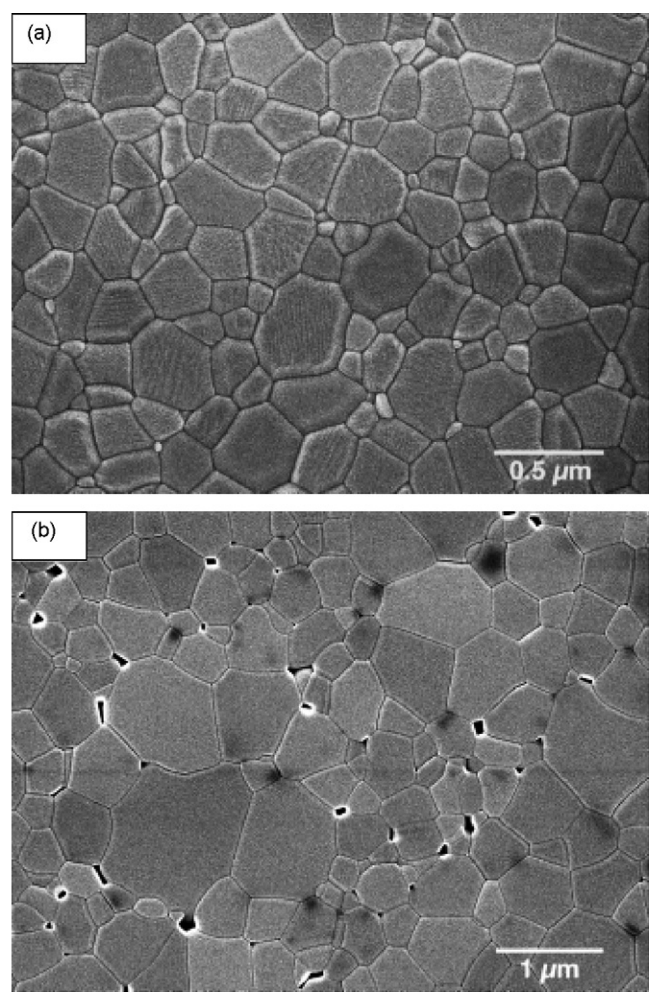

Fig. 2. Microstructures of the alumina sintered at a heating rate of (a) 2 °C/min and (b) 100 °C/min.