

CHANGES IN MICROSTRUCTURE OF SPHERICAL ALUMINA COMPACTS
DURING THE INTERMEDIATE AND FINAL STAGE OF SINTERING

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ABSTRACT

Changes in microstructure of spherical alumina compacts during the intermediate and final stage of sintering were studied by measuring the bulk density, grain growth, grain size and pore size distribution.

The relative density of 75-92% corresponds to the intermediate stage of sintering. The density increased linearly with logarithm of the soaking time, and the grain growth proceeded according to the equation of $a^3 - a_0^3 = kt$.

In the final stage of sintering, the porosity decreased linearly with logarithm of the soaking time, and the grain grew according to the equation of $a^2 - a_0^2 = kt$. The grain size distribution became to be almost a logarithm normal distribution. When the cavity just became to closed pore, the number and diameter of closed pore were depended on the initial particle diameter and the packing density of green compact. The pore size distribution did not change much in spite of the large grain growth, and the mean pore size kept nearly constant during the soaking. The number of pore decreased proportionally with logarithm of the soaking time.

1 INTRODUCTION

There are a lots of studies on the sintering of alumina, and the translucent or pore free alumina bodies were already manufactured. Now, control of microstructure of sintered body is the most significant theme in order to develop new substances, and many kinds of pure and fine alumina powder were synthesized by various methods [1]. Sintering process and microstructure of sintered body are strongly affected by the texture of green compact which is changed by shape, size and size distribution of particles. The quantitative studies of microstructure were mainly carried out by measuring the pore size distribution by means of a mercury penetrating porosimeter in initial and intermediate stage of sintering, and porosity change and kinetics of grain growth in the

intermediate and final stage of sintering [2-4]. But there are few studies to discuss grain and pore size distributions in the final stage of sintering [5].

In the previous study [6], author investigated for the changes in pore structure of compacts of various type of alumina powders during the initial stage of sintering by means of a mercury penetrating porosimeter. The pore diameter increased at first and then decreased in the fine powder compact in size of $0.3 \mu\text{m}$, but only decreased in the compact formed by pressing the spherical alumina in size of $12 \mu\text{m}$. In this study, microstructural changes of spherical alumina compacts during the intermediate and final stage of sintering were investigated by measuring the bulk density, grain growth, grain size and pore size distribution.

2 EXPERIMENTAL PROCEDURES

Spherical alumina particles were prepared by means of plasma flame method. After classified by elutriation, the spherical particles were annealed at 1300°C for 1 hour in order to transform to α phase [7]. Table 1 shows the range of elutriation and the mean diameter of the spherical alumina particles used in experiment. Figure 1 shows the particle size distribution of the sample (B).

The particles were pressed into compact at the pressure of $1000\text{kg}/\text{cm}^2$. And then the compact was fired at $1700\text{-}2000^\circ\text{C}$ for 0-16 hours in the vacuum of $5 \times 10^{-6}\text{mmHg}$. The relative density of the green compact was the range of 55-58%.

Microstructure of the fractured surface of sintered body was observed, and grain and pore size distributions were measured in the polished surface of the samples.

Table 1 Range of elutriation and mean diameter of the spherical alumina particles used in experiment

Sample No.	Range of elutriation	Diameter at 50 weight %
A	under $10 \mu\text{m}$	$8.4 \mu\text{m}$
B	$4 \mu\text{m} - 6 \mu\text{m}$	$5.3 \mu\text{m}$
C	$6 \mu\text{m} - 8 \mu\text{m}$	$6.8 \mu\text{m}$
D	$8 \mu\text{m} - 10 \mu\text{m}$	$8.9 \mu\text{m}$
E	$10 \mu\text{m} - 15 \mu\text{m}$	$11.7 \mu\text{m}$
F	$15 \mu\text{m} - 20 \mu\text{m}$	$16.0 \mu\text{m}$

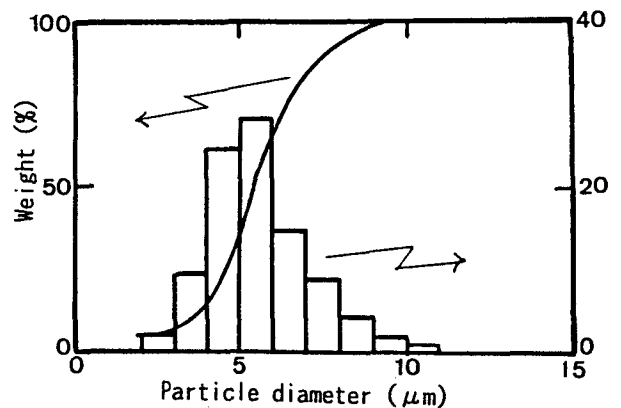


Fig.1 Particle size distribution of the sample (B)

3 RESULTS AND DISCUSSION

Figure 2 shows the time dependence of bulk density of the compacts sintered at 1700°C. The range of relative density was 65-88%, which is considered to correspond to the intermediate stage of sintering. Figure 3 shows the correlation between the relative density and logarithm of the soaking time. The linear relation was observed. The slope was not a constant value but changed to a small value with increasing of the particle size.

The particle kept sphere shape and grew little before the relative density became to be 75%. After the relative density reached to 75%, the particle changed into the polyhedron consisted from the surfaces of tetragon-heptagon, and it grew to considerably large size. Open pores existed among the grain boundaries, and they connected each other. The shape of the pore was not a cylinder which was assumed by Coble [3], but a polypillar or a polyhedron connected with the other pores. Closed pore was not observed even in the sample (B) soaked for 16 hours of which the relative density was 88%. Closed pores were obviously observed in the samples sintered at the higher temperature, where the relative density was about 92-93%.

For grain growth, the following equation was obtained in the intermediate stage; $a^3 - a_0^3 = kt$, where, a is the mean diameter of the grain at the soaking time t , a_0 is the mean diameter of the initial particles, k is the constant. This relation was often observed for sintering process of alumina [3].

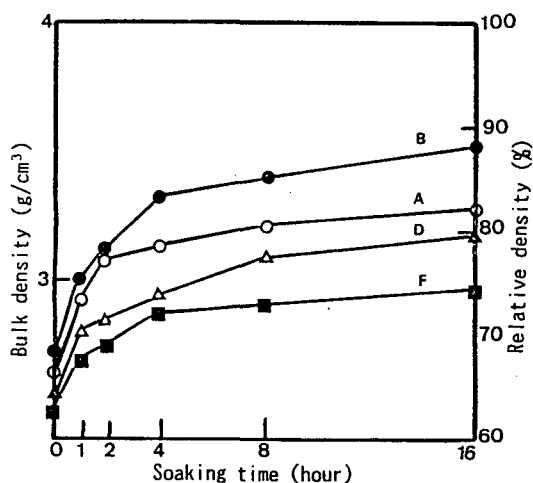


Fig.2 Time dependence of the bulk density of the compacts sintered at 1700°C

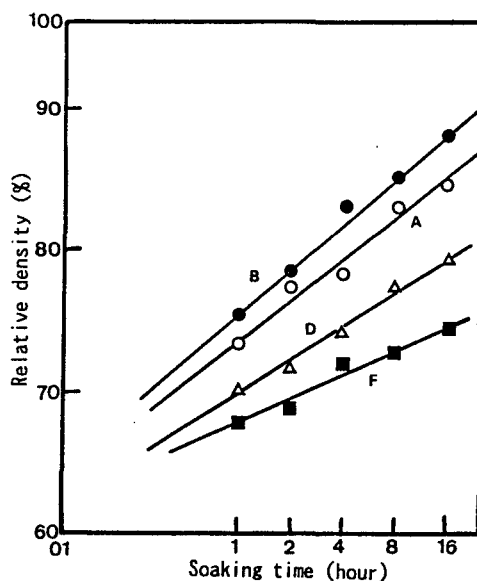


Fig.3 Correlation between the relative density and logarithm of the soaking time

Figure 4 shows the time dependence of bulk density of the compacts sintered at 1900°C. The range of relative density of the samples (A) and (B) was above 92%, which is considered to correspond to the final stage of sintering.

The grain grew over 75 μ m in diameter, but discontinuous grain growth did not observed. The closed pores with polyhedron shape existed among the coner of grains at first, then the most of them was included in the grains due to the large grain boundary migration. The pores changed to spherical shape and decreased in size with the soaking time. But a part of pores existed among the grain boundaries, and kept large and nonspherical shape.

Figure 5 shows the time dependence of the apparent grain diameter of polished surface of the sample (B) sintered at 1900°C. The values of diameter were measured by the largest distance across the grain for constant direction in the polished surface. The value of peak top was about 5 μ m, and the distribution was very narrow at 0 hour. According to the soaking time, the peak top shifted to a large value and the distribution became wide. The distribution curve was not symmetrically but spread to large size. The same results were observed in the other samples. Figure 6 shows the plots of distribution curves vs logarithm of the apparent grain diameter. The logarithm normal distribution curve was observed for the apparent grain diameter of all the samples. The actual grain size distribution seemed to change as like the apparent distribution.

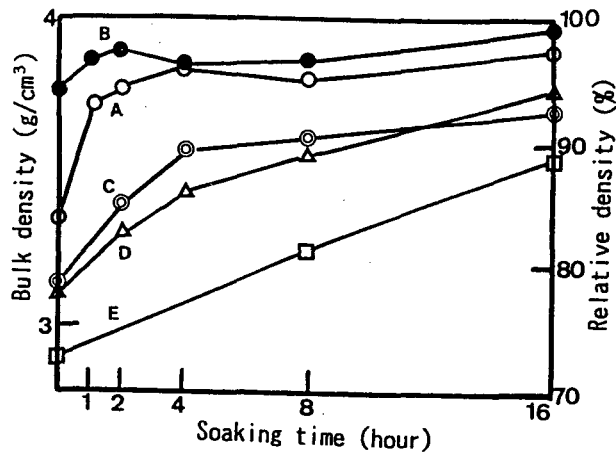


Fig.4 Time dependence of the bulk density of the compacts sintered at 1900°C.

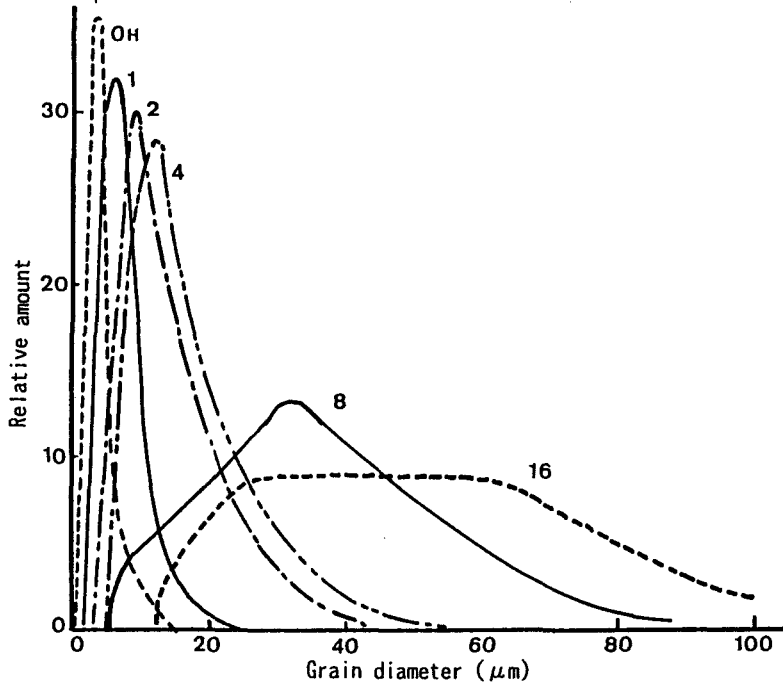


Fig.5 Time dependence of the apparent grain diameter of the polished surface of the sample (B) sintered at 1900°C

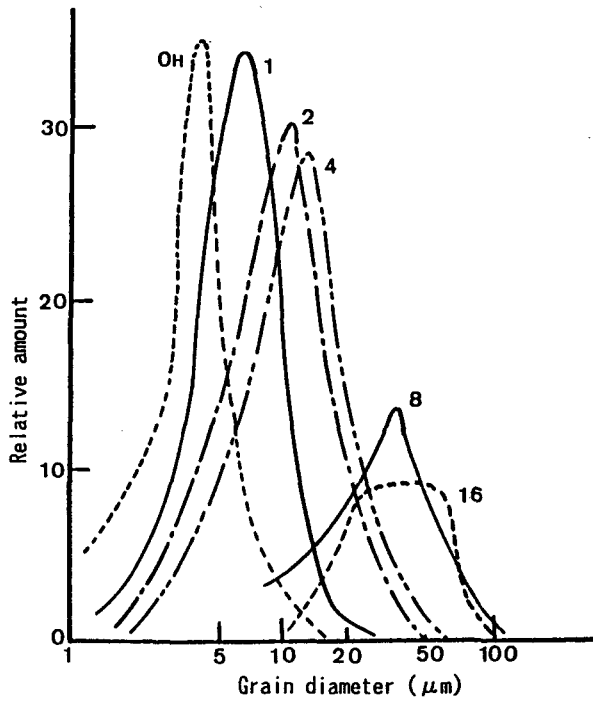


Fig.6 Plots of distribution curves vs logarithm of the apparent grain diameter

Figure 7 shows the correlation between logarithm of the mean grain diameter and logarithm of the soaking time of the samples sintered at 1900 °C. The linear relation was observed and the slope was 1/2. The relation of $a^2 - a_0^2 = kt$ was obtained, which changed from that of the intermediate stage of sintering. It is not clear for the reason of difference of the time dependence of grain growth between the intermediate and the final stage of sintering.

Figure 8 shows the distributions of pore diameter of the sample (B) sintered at 1900 °C. The pore diameter was smaller than 8 μm, more than a half of the pore was smaller than 2 μm, and the distribution did not change much with soaking time, though the large grain growth was observed. The mean diameter of pore kept nearly constant during the soaking time.

Figure 9 shows the distributions of pore diameter of the various samples sintered at 1900 °C for 16 hours. There were the larger pores in the samples, formed by pressing the larger spherical particles.

Figure 10 shows the correlation between the mean diameter of pores in the sintered sample and mean diameter of spherical particles of the starting compacts. The linear relationship was observed between both diameters. The dotted line in the figure shows relationship between the calculated mean diameter of cavities and the mean diameter of starting spherical particles. The calculated diameter of cavity was obtained from the equation of $d = (2-1)a_0$ which was derived from the initial diameter of particle and packing density of the compact. The relationship between pore diameter

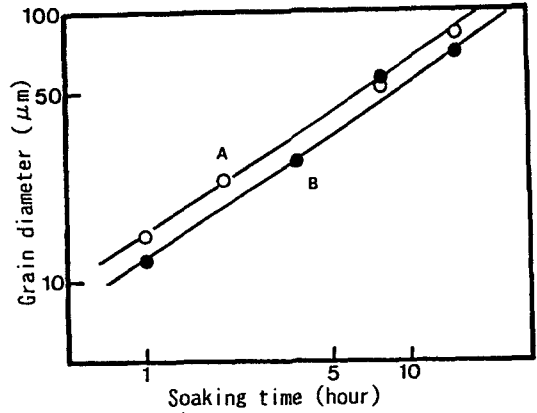


Fig.7 Correlation between the logarithm of mean grain diameter and logarithm of the soaking time of the sample sintered at 1900 °C

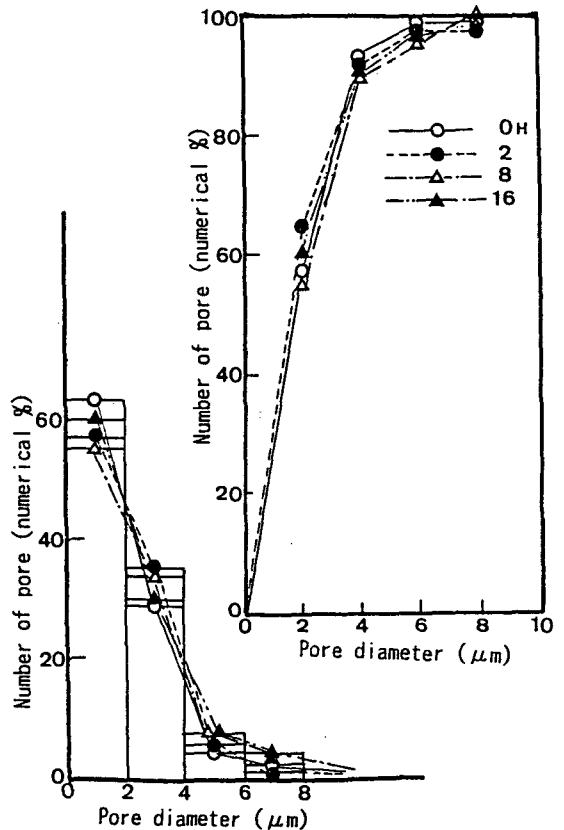


Fig.8 Distributions of pore diameter of the sample (B) sintered at 1900 °C.

and diameter of spherical particle obtained from experiments was the almost same to the calculated relationship, it was suggested that the cavity among particles became to the closed pore nearly maintaining the initial size.

In the initial stage of sintering, the pore size distribution was measured by means of mercury penetrating porosimetry. The result showed that the pore diameter decreased with sintering process [6]. This result seemed to be disagree with the result precedingly mentioned. The difference between them was caused by the methods of measurement. The pore diameter measured by mercury penetrating method shows the value of the entrance of open pore which is smaller than the actual pore diameter.

Figure 11 shows the correlation between the porosity and the number of pore in the area of $9 \times 10^4 \mu\text{m}^2$ of the polished surface of the samples sintered at 1900°C , and logarithm of the soaking time. In this figure, the data at 0 hour were plotted on the position at 0.01 hour. About 700 of pores existed in the area of $9 \times 10^4 \mu\text{m}^2$ of the polished surface of the sample (B) sintered for 0 hour, when the cavity just became to closed pore. The number corresponds to $8 \times 10^9/\text{cm}^2$, and it was agreed with the calculated number of $5 \times 10^9/\text{cm}^2$. Porosity and the number of pore decreased proportionally with logarithm of the soaking time. The porosity decreased with decreases of the number of pore. These results suggested pore behavior during the intermediate and the final stages sintering.

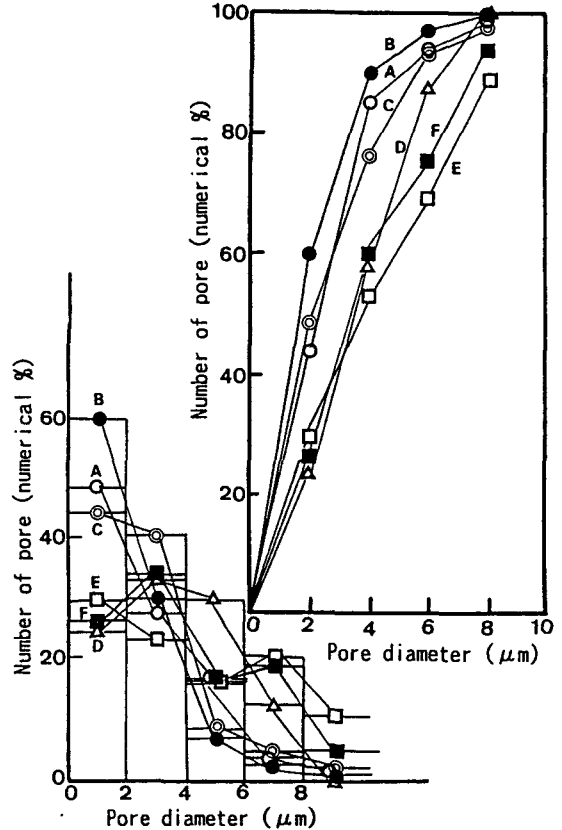


Fig.9 Distributions of pore diameter of the various samples sintered at 1900°C for 16 hours

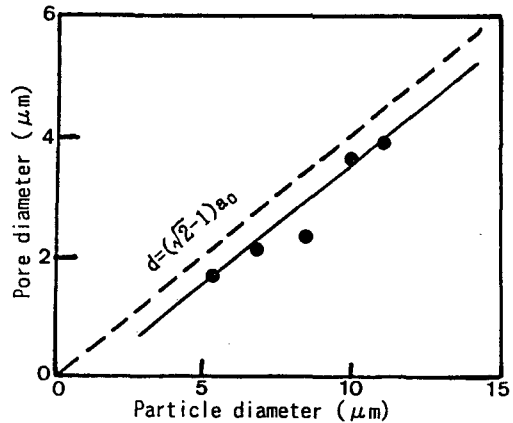


Fig.10 Correlation between the mean diameter of pores in the sintered sample and mean diameter of spherical particles at the starting compacts.

That is, pores did not move with grain boundary migration and did not associate with each other, and the diameter of each pore decreased with time and then the small pore disappeared. On the other hand a part of large pore acted as a vacancy sink and nearly kept the diameter, and totally the pore size distribution did not change so much and the number of pore decreased with the soaking time.

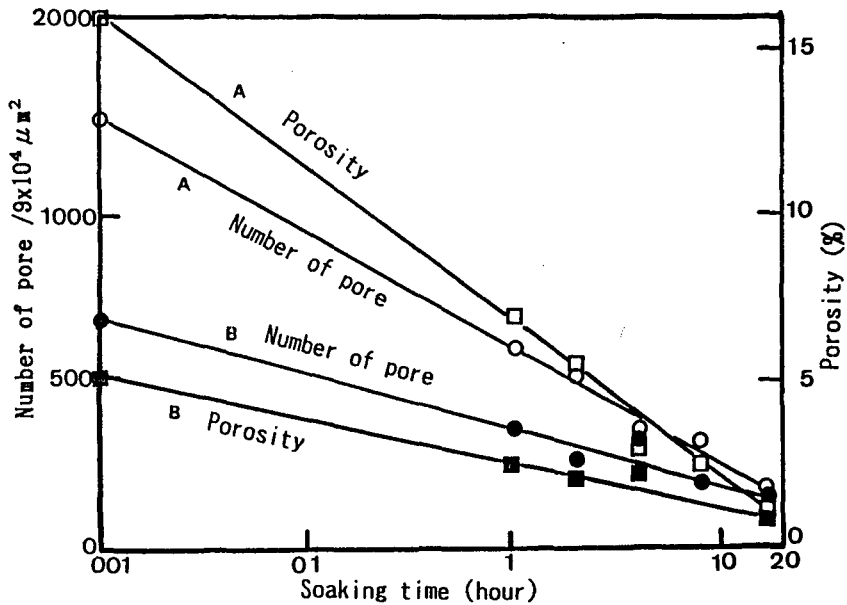


Fig.11 Correlation between the porosity and the number of pore in the area of $9 \times 10^4 \mu\text{m}^2$ of the polished surface of the samples sintered at 1900°C and logarithm of the soaking time.

4 CONCLUSIONS

Microstructural changes of spherical alumina compacts during the intermediate and final stage of sintering were studied by measuring the bulk density, grain growth, grain size and pore size distribution. The results were obtained as follows;

(1) The relative density of 75-92% corresponds to the intermediate stage of sintering. The relative density increased linearly with logarithm of the soaking time and the grain growth proceeded according to the equation of $a^3 - a_0^3 = kt$.

(2) In the final stage of sintering, the porosity decreased linearly with logarithm of the soaking time, and the grain became larger and the distribution spread to wide with the soaking time.

- (3) The grain grew according to the equation of $a^2 - a_0^2 = kt$, and the grain size distribution became to be almost logarithm normal distribution.
- (4) When the cavity just became to closed pore, the number and the diameter of closed pore depended on the particle diameter and the packing density of green compact.
- (5) The pore size distribution did not change much in spite of the large grain growth, and the mean pore size kept nearly constant during the soaking.
- (6) The number of pore decreased proportionally with logarithm of the soaking time.

5 REFERENCES

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