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Original research paper

Influence of coarse particles on microstructure of aluminum nitride sintered body

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ABSTRACT

Aluminum nitride (AlN) is used for quick diffusion and elimination of heat that is generated from electronic devices, such as power modules used for hybrid cars and micro processing units (MPUs) of computers. AlN provides high thermal conductivity, and it is known that its sintering performance and sintered body characteristics vary with the quality of AlN raw powder. When two types of commercially available AlN raw powder produced by the same reductive nitriding method were compared, the sintering performance and the thermal conductivity of sintered compacts processed from low-price AlN material powder were found to be lower than those of sintered compacts processed from high-price, high-purity, evenly granulated, and fine AlN material powder. As one of the causes of the foregoing, the effect of coarse particles contained in AlN material powder was investigated. The investigation results indicated that the coarse particles were AlN and the powder with the coarse particles removed by sifting out with sieves provided sintering performance and sintering behavior similar to those of high-price and high-purity AlN material powder. It was therefore found that the coarse particle constituted a sintering inhibiting factor. This paper reports the investigation results.

1. Introduction

With a recent increase in the capacity and speed of information processing, the operation frequency of MPU (micro processing unit) used for computers has been remarkably increased, and an increase in calorific value causes a big issue. Furthermore, in the field of electric power control, from the viewpoint of global environment conservation, development of electrically powered cars and hybrid cars is accelerated, and as a result, reduced size and increased efficiency of a power module, main component, have been carried forward. In such event, too, an increase in the calorific value causes an issue.

Because aluminum nitride (AlN) is the material that provides high thermal conductivity and high insulation properties, it is suited for the substrate that quickly diffuses and eliminates large heat generated in MPU of computers, power modules, and others [1–3]. For material powder for manufacturing aluminum nitride (AlN) sintered compacts, powder produced by three kinds of synthesis methods, namely, reductive nitriding, direct nitriding, and chemical vapor deposition are commercially available, but the reductive nitriding powder is most popularly manufactured and put on market. The reason this powder is used is that it is possible to obtain evenly granulated, highly pure, comparatively low-price raw powder.

When the following two types of AlN raw powder produced by the same reductive nitriding method were compared, the two types of powder are distinguished as described below because of the difference in the grain size of aluminum oxide (Al_2O_3) of raw material in each type, or the difference in purity or the difference in process between AlN raw powder ① and ②. ① is evenly granulated, highly pure and high-price raw powder, and AlN raw powder ② is raw powder with lower purity containing coarse particles. The sintering performance of AlN raw powder ① is higher, and sintered compacts with high thermal conductivity can be obtained easily [4–7]. For the causes, various factors are considered, such as grain sizes of AlN raw powder, grain size distribution, the content of oxygen, metals and other impurities as well as coarse particles contained in AlN raw powder, strain in AlN particles, or the like, but we assumed that one of the causes of differences in sintering behavior generated particularly between reductive nitriding powder results from the coarse particles contained in AlN raw powder ②, which most probably obstruct the sintering of the powder in the process, and we have made studies on the influence of coarse particles on the sintering performance [8,9].

This report clarifies the influence of the coarse particles on the sintering process of AlN by determining the real identity of coarse particles through assessment of optical characteristics by the use

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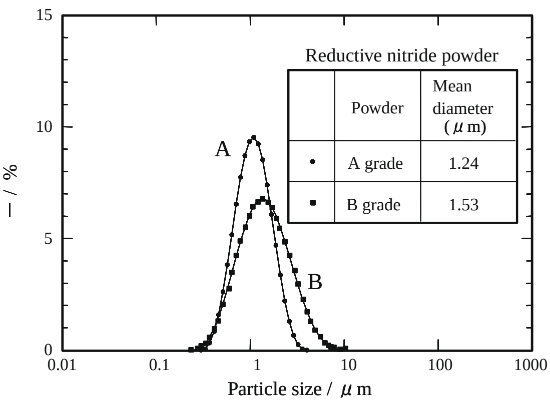

Measuring apparatus : Mastersizer 2000
Solvent : ethylene glycol

Fig. 1. The distribution of AlN material powder.

of polarizing microscope on the basis of analysis of crystallinity of coarse particles, crystal orientation of fine crystals, etc. contained in low-cost AlN powder (reductive nitriding powder) that contains a large quantity of coarse particles, furthermore by sintering by the use of the AlN powder with coarse particles eliminated by sieving as material, which are considered to obstruct the sintering of AlN from raw powder, and by comparing them with sintering phenomena of AlN material powder ①.

2. Methods

2.1. Analysis of rough particles in raw powder

Two types of AlN powder, which were produced by the same reductive nitriding method and which were different in particle size distribution, were used (i.e., Tokuyama-made A-grade powder and B-grade powder). Fig. 1 shows the particle size distribution of each type of powder. These types of AlN powder were weighed and sampled, and 150 g each of the samples was added into 500 ml of ethanol manufactured by Junsei Chemical Co., Ltd., exposed to ultrasonic waves for 10 min, and dispersed. The slurry of each sample obtained was classified by a wet classification method with a circular vibrating sieve (VSS-50 compact vibrating shaker) and nylon sieve (with mesh opening sizes of 5, 8, 10, and 20 μm) both manufactured by Tsutsui Rikagaku Kikai Co., Ltd.

From the weight change of each sieve before and after the above classification, the weight of the residual powder on the sieve was measured and the weight fraction of the powder sample at each class was calculated. Furthermore, a scanning electron microscope (JSM-5200 SEM) manufactured by JEOL Ltd. and a polarization microscope (ECLIPSE E600 POL) manufactured by Nikon Corporation were used to observe the form of the powder sample at each class. For the observation of the form with the polarizing microscope, in particular, diiodo methane manufactured by Junsei Chemical Co., Ltd. was used for an immersion liquid. Then the samples were observed under an open nicol and crossed nicol for the evaluation of the optical anisotropy of the particles.

Furthermore, the constituent phase of each powder sample was identified by an X-ray diffraction method. Then, with the segregation of coarse particles and agglomerated particles consisting of ultra-fine grains based on SEM photographs, the existence ratio of coarse particles was obtained. As a result, the content rates of coarse particles in A-grade powder and B-grade powder were calculated from the weight fraction and the existence ratio of coarse particles in each powder sample.

2.2. Influence of coarse particle removal on sintering of AlN

A sintering experiment was conducted by using raw B-grade powder and classified B-grade powder that was filtered through a sieve with a mesh opening size of 5 μm. Then Y_2O_3 powder was added at a ratio of 5% by mass to each sample. Then a total of 50 g of powder was wet blended in ethanol. After the ethanol was evaporated and removed, paraffin (at a melting point of 46–48 °C) manufactured by Junsei Chemical Co., Ltd. was added at a ratio of 1% by mass. Again, the sample was filtered through a sieve with a mesh opening size of 0.25 mm by force to obtain granules. This powder was uniaxially molded at 60 MPa and CIP molded at 200 MPa, and a green body ($\phi 15 \times 5$ mm in size) was obtained. The green body was degreased and located in an AlN crucible set in a graphite crucible. Then the graphite-made crucible was set in a carbon heater, which finally became sintered in a horizontal-type high-frequency induction heating furnace at a sintering temperature of 1650–1800 °C for a soaking time of 2 h at the N_2 flow rate of 1 l/min.

3. Results and discussion

Table 1 shows the weight fraction of residual particles on each sieve. This showed that there were a few A-grade and B-grade powder particles on the sieves. They may have been AlN coarse particles, agglomerated particles consisting of AlN ultra-fine grains, impurity particles, such as rubber, glass, or plastic particles, mixed in the process. Therefore, the residual particles remaining on the sieves were identified.

Fig. 2 shows the SEM photographs of the residual particles on each sieve. From this, it has become clear that most of the residual particles of A-grade powder on the sieves were agglomerated particles consisting of ultra-fine grains. On the other hand, most of the residual particles of B-grade powder on the sieves were coarse particles with a diameter range of several to several tens of microme-

Table 1
The containing rate of coarser grains in each powder.

Range (μm)	Weight rate	Weight rate around grain size 1 μm (A)	Existence rate of coarser grains (B) (%)	Containing rate of coarser grains A × B
A-grade powder				
0–5	98.86%	–	0	0
5–8	3700 ppm	1233 ppm/μm	0	0
8–10	1400 ppm	700 ppm/μm	0	0
10–20	4800 ppm	480 ppm/μm	21.1	101
~20	1500 ppm	–	0	–
B-grade powder				
0–5	99.55%	–	0	0
5–8	1900 ppm	633 ppm/μm	31.6	200
8–10	600 ppm	300 ppm/μm	78.6	235
10–20	1400 ppm	140 ppm/μm	88.2	123
~20	500 ppm	–	0	–