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EFFECT OF PARTICLE SIZE DISTRIBUTION ON THE MICROSTRUCTURE AND MAGNETIC PROPERTIES OF SINTERED NdFeB MAGNETS

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Abstract. Effects of mean powder size and size distribution on the microstructure and magnetic properties of anisotropic NdFeB sintered magnets were investigated. The hydrogen-decrepitated NdFeB alloy was jet milled and the jet-milled powder was subsequently divided by a classifier in order to eliminate coarse and fine particles. The classified powder was then compacted, sintered, and annealed at certain conditions to make anisotropic sintered magnets. The maximum energy product of the sintered magnet improved with the removal of fine particles while the coercivity of the magnet increased effectively by eliminating coarse particles. However, when the magnet was made with the powder mostly consisting of fine particles (mean particle size = 3.35 μm), the coercivity dropped significantly due to extreme abnormal grain growth caused by too high sintering temperature and too much oxygen content in the powder.

1. INTRODUCTION

Since the NdFeB magnets were first introduced in 1984 [1,2], there have been extensive studies due to their outstanding magnetic properties. The recent issue on these magnets, especially for anisotropic sintered magnets, is how to improve coercivity of the magnets effectively without using too much heavy rare earths so that the magnets can be used in HEV and EV [3]. In general, magnetic properties of a NdFeB sintered magnet depend on its microstructures predetermined by processing steps such as strip casting, hydrogen treatment, jet milling, compaction, sintering, and post sintering [4]. Each processing step is important and should be controlled carefully to improve the hard magnetic properties. In powder metallurgical process, characteristics of the powder is very important to determine the overall properties of final products. In this study, magnetic properties of NFeB sintered magnets are discussed in terms of the size and distribution of the classified jet-milled powder.

2. EXPERIMENTAL

The alloy with nominal composition of 29Nd-3.4Dy-1B-1TM-bal.Fe (wt.%) was prepared by a strip casting technique. The strip-cast alloy was decrepitated for 2 hours with a hydrogen pressure of 0.12 MPa. After hydrogen decrepitation, the alloy was dehydrogenated in a vacuum furnace and subsequently pulverized by jet-milling. To classify the jet-milled powder more precisely, a new classifier operated in the principle of centrifugal force was employed. The jet-milled powder was divided by the classifier into two different conditions: One at 5,800 rpm so that 16% of the classified powder to be fine powder (Powder I) and 84% of them to be coarse powder (Powder II), and the other at 2,700 rpm so that 81% of the classified powder to be fine powder (Powder III)

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Table 1. Mean particle size and particle size distribution of the starting powder and the classified powders.

Powders		Properties				
		Meanparticle size (μm)	D10 (μm)	D50 (μm)	D90 (μm)	D90/D10
Starting powder		4.14	2.66	4.29	6.05	2.27
1st condition (5800 rpm)	Powder I: fine particles	3.35	1.75	3.25	4.62	2.64
	Powder II: coarse particles	4.24	2.79	4.36	6.10	2.19
2nd condition (2700 rpm)	Powder III: fine particles	3.86	2.56	3.82	5.59	2.18
	Powder IV: coarse particles	5.17	3.72	5.40	7.33	1.97

Table 2. Magnetic properties and densities of the sintered magnets fabricated with the starting powder and the classified powders.

Used powders	Properties			
	Density (g/cm^3)	Magnetic properties		
		Br [kG]	iH_c [kOe]	$(BH)_{max}$ [MGOe]
Starting powder	7.62	13.1	19.0	42.2
Powder I	7.57	11.8	7.6	29.6
Powder II	7.58	13.3	19.2	43.2
Powder III	7.62	13.1	20.2	42.0
Powder IV	7.01	12.3	18.8	36.2

and 19% of them to be coarse powder (Powder IV). The classified powder was then pressed with a perpendicular magnetic field of 2 T. The green compacts were sintered in a vacuum furnace at 1060 °C for 4 hours, and then quenched to room temperature under Ar atmosphere. The sintered bodies were post annealed at 850 °C for 90 minutes and then at 500 °C for 2 hours.

Particle size distribution of the classified powder was measured with a particle size analyzer (API Aerosizer LD). Magnetic properties and oxygen content of the sintered bodies were measured with a BH loop-tracer and a LECO O_2/N_2 analyzer, respectively. Morphology of the powder and microstructure of the sintered body were investigated with a SEM (Hitachi N3000). Density of the sintered bodies was measured by using Archimedes' principle.

3. RESULTS AND DISCUSSION

Mean particle size and particle size distribution of the powder investigated in this study are summarized in Table 1. As we expected, the mean particle size of the classified powder became smaller when the classified speed was higher (1st condition). Accordingly, while the mean particle size of Powder II was slightly larger (4.24 μm) than that of the starting powder (4.14 μm), Powder IV that was classified at lower speed had much larger mean particle size of 5.17 μm. This size difference brought about the discrepancy in magnetic properties of the sintered magnets listed in Table. 2. It is noted that the mean particle size was slightly larger than D50 value for fine powder whereas that of the coarse powder including the starting powder was smaller than D50 (50 vol.% of particles have a size below the indicated value).

Fig. 1 shows morphology of the starting powder and the classified powder. It can be seen that the starting powder was a typical mixture of irregular shaped fine and coarse particles. After classification, particle size distribution became more uniform, especially for coarse powders (Figs. 1c and 1e). In fact, D90/D10 values (width of particle size distribution) of Powder II, III, and IV in Table 1 were all smaller than that of the starting powder. However, D90/D10 value of Powder I, possessing the smallest mean

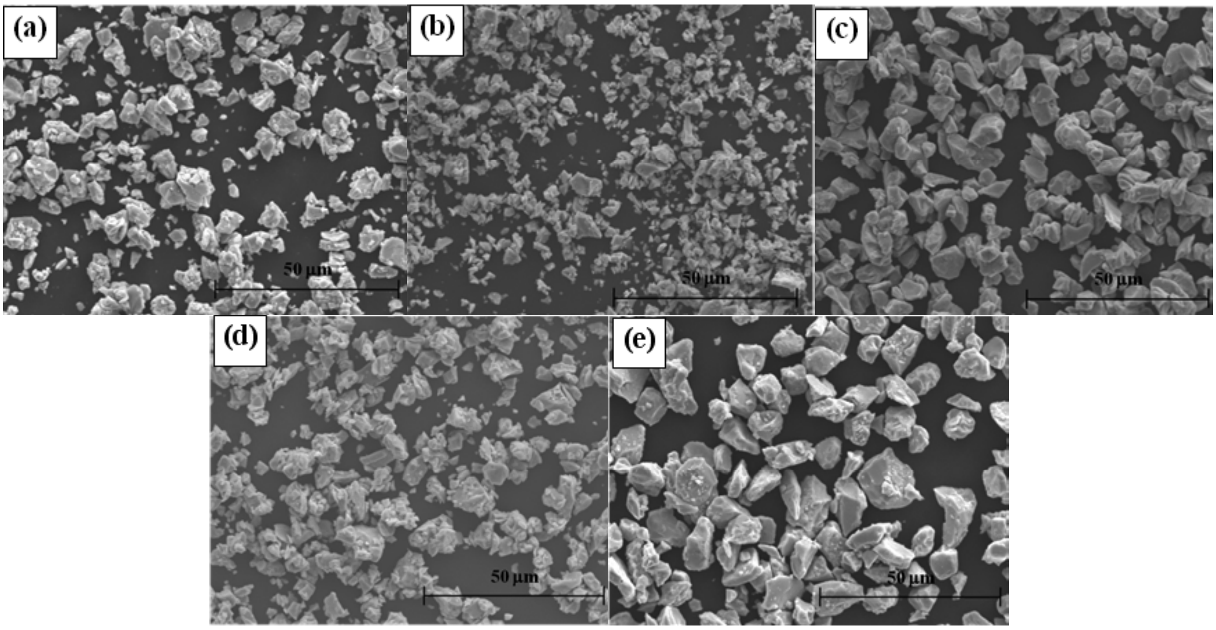

Fig. 1. SEM images of (a) starting powder, (b) powder I, (c) powder II, (d) powder III, and (e) powder IV.

particle size, was larger than that of the starting one, indicating that particle size distribution became worse when the powder consisted of very fine particles under this classification condition. Variation of mean particle size in Table 1 can be visually confirmed in this figure.

Magnetic properties and densities of the sintered magnets made with the starting powder and the classified powders are listed in Table 2, and the fracture surfaces of the sintered magnets are shown in Fig. 2. The density of a sintered body was higher when the magnet was made with the powder in which some fine particles were mixed with coarse particles properly as shown in Fig. 1. Therefore, while the magnets made with both the starting powder and Powder III exhibited full density, the magnets made with the powder mostly consisting of fine particles (Powder I) or coarse particles (Powder II) showed lower density values. In the case of Powder IV that has the largest mean particle size but the lowest D90/D10, i.e., the most uniform particle size distribution, the density was the lowest mostly due to many pores in the sintered body as shown in Fig. 2e. It tells us that sintering temperature to obtain full density has to be changed because the critical sintering temperature for full densification varies with particle size and distribution [5]. In general, the critical sintering temperature decreases with a fine powder, and there is about 50 °C difference when D50 value of the powder varies about 1.5 μm [5].

The remanence (B_r) of an anisotropic NdFeB magnet depends on magnetic volume, density, and grain alignment in the sintered magnet. For a given composition, therefore, improvement of grain orientation together with full densification is important for acquisition of high B_r. Previous researches showed that the degree of grain alignment is related to the particle size distribution [5,6]. There is a certain range of D10 and D90 which gives rise to better alignment depending on the microstructure of strip-cast alloy and average particle size. Fine powder usually decreases the degree of orientation since the torque on the small particles would be too low to overcome the friction between particles [6]. Comparing the results in Table 2 with those in Table 1, B_r of the magnets increased with the increase of the mean particle size, except for the one with Powder IV which had the lowest density due to many pores in the sintered body as shown in Fig. 2e. Accordingly, in spite of lower density, the magnet fabricated with the powder in which fine particles were mostly removed (Powder II) exhibited the high-

Table 3. Oxygen content of the sintered magnets fabricated with each powder.

Used powders	Oxygen content (ppm)
Starting powder	2082
Powder I	4325
Powder II	1642
Powder III	2087
Powder IV	1623

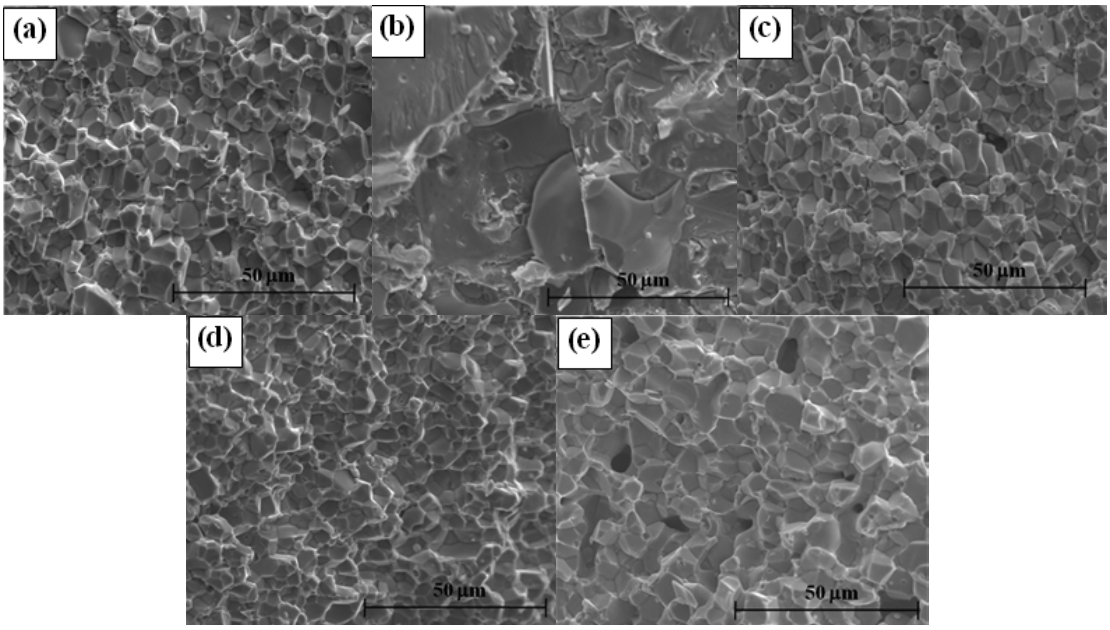

Fig. 2. Fracture surfaces of the sintered magnets fabricated with (a) starting powder, (b) powder I, (c) powder II, (d) powder III, and (e) powder IV.

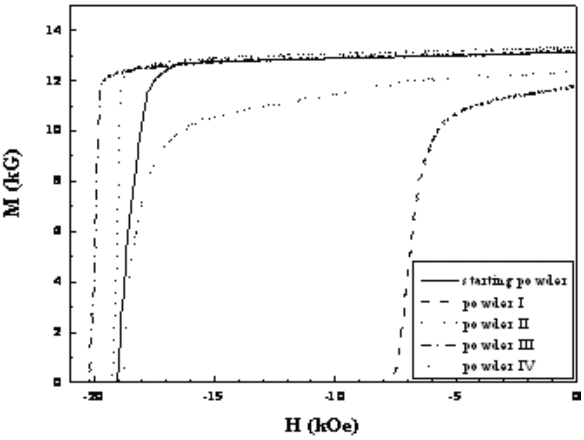

Fig. 3. Demagnetization curves of the sintered magnets fabricated with each powder.

est remanence and maximum energy product ($(BH)_{max}$), directly proportional to $(B_r)^2$, obviously due to better grain alignment.

The coercivity ($_iH_c$) of NdFeB sintered magnets is determined by the nucleation of reverse domains at the grain boundaries of the hard magnetic $Nd_2Fe_{14}B$ grains. Since smaller grain size reduces the probability of nucleation of reverse domains at the grain boundaries and local demagnetizing stray fields, the coercivity of sintered magnets usually shows a negative grain-size dependence [7]. That is, the coercivity of sintered magnets generally increases as grain size, i.e., particle size decreases. As shown in Table 2, the coercivity of sintered magnets increased with the decrease of grain size (See Fig. 2.), as well as mean particle size of the powder in Table 1. Considering the particle size, the highest coercivity should have been obtained from the magnet fabricated with the finest powder (Powder I). On the contrary, it showed the lowest coercivity because of extreme abnormal grain growth as revealed in Fig. 2b. The abnormal grain growth which could occur during liquid phase sintering is a thermally activated mechanism strongly dependent on the diameter of particles [8]. Below a critical diameter, the particles tend to minimize their surface energy, creating a nucleus of very large grain by the absorption of other particles.

Furthermore, as shown in Table 3, oxygen content in the magnets increased with the decrease of the mean particle size of the powders, exceeding 4,000 ppm for the one with Powder I, because the total surface area of the powder becomes larger as the particle size decreases further. After pulverization of hydrogen decrepitated strip-cast alloy, the jet-milled powder consists of $Nd_2Fe_{14}B$ single crystal particles that usually carry fragments of Nd-rich phase [4,7]. During liquid phase sintering, these Nd-rich fragments that have a low melting temperature play an important role for densification of a sintered body and isolation of $Nd_2Fe_{14}B$ grains, inhibiting unnecessarily large grain growth [9]. Since the Nd-rich phase is sensitive to oxidation, it is quite possible that such role of Nd-rich fragments for sintering can be reduced in the powder containing too much oxygen, which brings about low density and poor insulation of hard magnetic grains thereby an abnormal growth of the grains. Such abnormal grain growth shown in Fig. 2b reduces not only the coercivity but also the remanence because it causes random orientation of large grains [5].

Fig. 3. shows demagnetization curves measured from the sintered magnets fabricated with each powder. With the increase of magnetic properties as listed in Table 2, the squareness of demagnetization curves were improved when the jet-milled powder was classified properly (Powder II and III). However, if the powder was too fine (Powder I) or too coarse (Powder IV), the squareness became worse as the magnetic properties were deteriorated by the reasons described above.

4. CONCLUSIONS

Classification of the jet-milled powder had a beneficial effect on the improvement of magnetic properties of NdFeB sintered magnets. There was a certain range of particle size distribution (D90/D10) that brought about better magnetic properties. The remanence and maximum energy product of the sintered magnet usually improved with the removal of fine particles while the coercivity of the magnet increased by eliminating coarse particles. When the classified powder was too fine or too coarse, however, magnetic properties of the magnets were deteriorated due to extreme grain growth for the former and poor densification for the latter.

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