研究

Effects of Heat Treatment Temperature and Particle Size on the Coercive Force of Gas-atomized Nd-Fe-B Powder*

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Tsuguaki Ohki, Shoji Kanbe, Rikuo Ogawa and Nobuyasu Kawai: Effects of Heat Treatment Temperature and Particle Size on the Coercive Force of Gas-atomized Nd-Fe-B Powder.

Changes in the coercive force (H_c) of gas-atomized Nd-Fe-B powder were measured as functions of annealing temperature and particle size. The H_c of the powder (sieved to a particle size of $<44\ \mu\text{m}$) increased with increasing annealing temperature, reached a maximum at 600°C and then decreased. In the H_c particle size dependence curves, the maximum levels of H_c of as atomized and annealed powders at 600°C were observed in the $60\sim 90\ \mu\text{m}$ and $10\sim 20\ \mu\text{m}$ diameter size ranges, respectively. The $10\sim 20\ \mu\text{m}$ diameter powder annealed at 600°C for two hours exhibited an H_c of more than $10\ \text{KOe}$. The experimental results indicated that the H_c of Nd-Fe-B gas-atomized powder was dependent on strain and grain size as well as on the structure of the matrix phase and grain boundary. (Received February 3, 1988)

I Introduction

In recent years, considerable attention has been given to permanent magnets based on Nd-Fe-B alloys, which have been shown to possess outstanding magnetic properties^{1~5}). A great deal of work has been done on the Nd-Fe-B sintered magnet, but very few studies have been reported on the hard magnetic properties of fine powders made out of these alloys^{6,7}).

The present report is concerned with the effects of heat treatment temperature and particle size on the coercive force of gas-atomized Nd-Fe-B powder.

II Experimental procedures

The alloy having a nominal composition of 27.71 wt%Nd, 1.41 wt%B, 6.81 wt%Co, and 3.44 wt%Dy, bal. Fe was induction melted under an Ar atmosphere. The ingot was remelted and atomized using the Ar gas atomization process. In Ar gas atomization the size distribution of particles collected can extend from below $10\ \mu\text{m}$ up to a few hundred μm , depending on the ratio of gas flow to metal melt flow and on the nozzle diameter.

The atomized particles were sieved, using a turbo classifier in a N_2 gas atmosphere, and sorted into two categories: first, $0\sim <44\ \mu\text{m}$, and second, into the separate ranges of $3\sim 6$, $6\sim 10$, ..., $210\sim 250\ \mu\text{m}$. The powders were heat-treated in evacuated quartz tubes at temperatures of $400^\circ\text{C}\sim 1050^\circ\text{C}$ for 2 hours and $500^\circ\text{C}\sim 900^\circ\text{C}$ for $0.5\sim 9$ hours. The magnetization was measured on these samples, using a vibrating sample magnetometer (VSM) with a maximum magnetic field of $15\ \text{KOe}$. In order to observe the shape of the powders and the phases appearing in the powders, a metallogra-

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phical investigation was made using optical microscopy and a scanning electron microscopy (SEM). The X-ray diffraction measurements were carried out on the powders by means of Cu-K α radiation and a graphite crystal monochromator.

III Results and Discussion

The particle size distribution of the atomized Nd-Fe-B powders is shown in Fig. 1. The average size of the powders was 36 μm . A SEM photograph of the atomized powders is shown in Photo. 1. As seen in the photo, each powder exhibits a spherical shape, and the surfaces of the particles are relatively smooth.

The experimental results of Hc as a function of heat treatment temperature are shown in Fig. 2 for the powders sieved to a particle size of <44 μm . As the temperature rose, Hc began to increase from 500°C, reached a maximum value at 600°C~700°C, and then decreased slightly until around 1000°C. At still higher temperatures the Hc went into a precipitous drop.

Fig. 3 shows the variation of the X-ray diffraction powder spectra with increases in the annealing temperature for diffraction angles between 37 and 40.5 degrees.

The peaks in the figure correspond to the high points of intensity which were present in the tetragonal Nd₂Fe₁₄B type phase. With increases in the annealing temperature, a different refraction appeared and the intensity of the Nd₂Fe₁₄B peaks increased. The different phase seems to be the Nd₂Fe₇B₆ phase. The effect of the annealing on the Hc of Nd-Fe-B magnet has been ascribed to grain size, structure of grain boundary and strain.

In Fig. 2 the increase in Hc with annealing is seen to associate with the release of strain in the Nd₂Fe₁₄B phase and with the precipitation of a phase in which the powder apparently consists of Nd₂Fe₇B₆. At higher temperatures, where the Hc dropped off, the grain size became larger.

Fig. 4 shows the measurement of Hc in as-atomized and annealed powders as a function of particle size.

The Hc of the powders annealed at 600°C increased for particles in all size ranges. The Hc of as-atomized powders increased with the particle size up to particles of about 60~90 μm , reached a maximum value of about 4 KOe, and then decreased once more. In contrast, the Hc of powders annealed at 600°C for 2 hours also increased with particle sizes of up to 10~20 μm , reached a maximum value of about 11 KOe, and then decreased gradually.

Photo. 2. shows the microstructures of powders with different particle sizes. In proportion to

Fig. 1. Particle size distribution of gas-atomized Nd-Fe-B alloy powders.

Photo. 1. Morphology of gas-atomized Nd-Fe-B alloy powders.

Fig. 2. Variation of H_c with annealing temperature for Nd-Fe-B alloy powders.

Photo. 2. Optical micrograph of Nd-Fe-B alloy powder sieved to particle sizes of (a) $<44\mu\text{m}$, (b) $44\sim77\mu\text{m}$, (c) $>77\mu\text{m}$.

Fig. 3. X-ray diffraction spectra of gas-atomized Nd-Fe-B alloy powders sieved to a particle size of $<44\mu\text{m}$, and annealed at different temperatures.

(a) as atomized, (b) $500^\circ\text{C}\times 2\text{h}$,
(c) $600^\circ\text{C}\times 2\text{h}$, (d) $900^\circ\text{C}\times 2\text{h}$.

Fig. 4. Variation of H_c with particle size of as-atomized Nd-Fe-B alloy powders and those annealed at 600°C for 2 hours.

Fig. 5. Variation of microstructure size with particle size in the gas-atomized Nd-Fe-B powders.

the particle size, the microstructure in the powders grew bigger.

In Fig. 5 the measured size of the microstructures of the powders is plotted against the particle size for as-atomized and annealed powders. A linear dependence can be seen between the size of microstructure and the particle size. The data seem to show that the reason for the decrease of the H_c at the larger particle sizes is the larger sizes of the microstructure.

Fig. 6 shows the X-ray diffraction patterns of as-atomized powders with the different particle sizes. The patterns for the smaller particle sizes correspond to the $\text{Nd}_2\text{Fe}_{14}\text{B}$ phase only.

As the particle sizes increased, the powder went through different phases and the diffractions seen in the $\text{Nd}_2\text{Fe}_{14}\text{B}$ phase were considerably sharpened. On the basis of these data, it seems reasonable to conclude that the low H_c at the smaller particle size was due to residual strain. The cooling rate for the smaller powders was higher than that for the larger powders. Therefore Nd and B atoms were dissolved and supersaturated into the $\text{Nd}_2\text{Fe}_{14}\text{B}$, and the strain was retained. The reason that the H_c for the smaller annealed powders showed a low value is the fact that the strain was not completely released with annealing at 600°C for 2 hours.

The proof of this is the fact that the different phase can not be seen in the annealed powders with smaller particle sizes, as shown in the first two spectra in Fig. 7.

It is generally believed that the H_c in hard magnetic materials is influenced by particle size, strain, structure of matrix phase and grain boundary. The data seem to indicate that the change in the H_c of Nd-Fe-B atomized powders is mainly related to changes in the first two factors, that is, particle size and strain. The H_c decreases with increases in particle size. As a consequence of annealing, the strain leaves the powder, resulting in an increase in H_c .

In summary, then, the data resulting from these experiments are fairly conclusive. They would seem to indicate that limiting the size of powder particles and including of annealing in the process of powder production improve the magnetic properties of such alloy powders as Neodimium, Iron and Boron, leading to better magnets.

Fig. 6. X-ray diffraction spectra of gas-atomized Nd-Fe-B alloy powders sieved to particle sizes of

- (a) 3~6 μm , (b) 20~44 μm ,
(c) 44~77 μm , (d) 105~144 μm .

Fig. 7. X-ray diffraction spectra of gas-atomized Nd-Fe-B alloy powders sieved to particle sizes of (a) 3~6 μm , (b) 20~44 μm , (c) 105~144 μm and annealed at 600°C for 2 hours.

Reference

- 1) M. Sagawa, S. Fujimura, N. Togawa, H. Yamamoto and Y. Matsuura: J. Appl. Phys., **55** (1984), 2083.
- 2) J. J. Croat, J. F. Herbst, R. W. Lee and F. E. Pinkerton: J. Appl. Phys., **55** (1984), 2078.
- 3) G. C. Hadjipanayis, R. C. Hazelton and K. R. Lawless: J. Appl. Phys., **55** (1984), 2073.
- 4) N. C. Koon and B. N. Das: J. Appl. Phys., **55** (1984), 2063.
- 5) K. S. L. Narashimhan: J. Appl. Phys., **57** (1985), 4081.
- 6) R. W. Lee: Appl. Phys. Lett., **46** (1985), 790.
- 7) K. Gudimetta, C. N. Christodoulou and G. C. Hadjipanayis: Appl. Phys. Lett., **48** (1986), 670.