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Development of Nd-Fe-B Anisotropic Bonded Magnets

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Abstract:

A packed rolling process was proposed for making a rapidly quenched Nd-Fe-B powder anisotropic, and an anisotropic Nd-Fe-B powder with excellent magnetic properties was successfully produced on an industrial scale. Technology was also established for compacting the anisotropic Nd-Fe-B powder in a magnetic field. Radially-oriented anisotropic ring magnets, the most widely used type of shape, were produced with the maximum energy product $(BH)_{max}$ of 16 MGOe, which proved their superiority to conventional bonded magnets in on-circuit evaluation in motors.

1. Introduction

Rare earth magnets have rapidly grown in market size since the 1983 invention and commercialization of neodymium-iron-boron (Nd-Fe-B) magnets. This is because the development coincided with the high-growth period of the electronics industry. As of 1991, the production of sintered magnets including samarium-cobalt (Sm-Co) magnets recorded ¥52.2 billion, and that of bonded magnets registered ¥9 billion[1]. A recent trend is the growth of bonded magnets in the whole rare earth magnet production. This is because isotropic resin-bonded magnets produced from rapidly quenched Nd-Fe-B powders have come to be used in large quantities, mainly for small-sized motors. **Fig. 1** shows the main shapes and applications of rare earth magnets. Bonded magnets are deprived of magnetization by the resin content, but are suited for thin and small components. To be noted in this connection is that radially anisotropic and thin magnets are difficult to make by the sintering process.

Bonded magnets are produced by compacting magnet powders with a resin binder. Nd-Fe-B magnets now in use are produced by mixing the Nd-Fe-B powder prepared by melt spinning with the binder and compression-moulding the mixture. Therefore, they are magnetically isotropic, and their maximum energy product $(BH)_{max}$, an important index of magnet properties, is 8 to 10 MGOe.

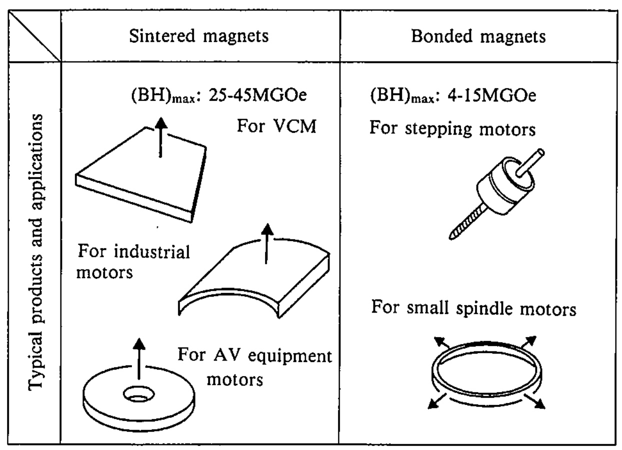

Fig. 1 Major shapes and applications of rare earth magnets

The recent need for reducing the size of electronic equipment demands bonded magnets with higher magnetic properties. To meet this requirement, the Nd-Fe-B powder is rendered anisotropic, magnetically aligned in a magnetic field, and compression-moulded. A combination of hot pressing and die upsetting[2,3] used to be employed to make magnet powders anisotropic, but this process has many problems to be solved. The authors proposed a packed rolling process to more economically obtain anisotropic powders[4]. An early problem of thermal instability

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was solved by adding copper. Their studies also dealt with the magnetic alignability[5] of magnet powders and the magnetic properties[6] of bonded magnets.

This article introduces the results of a series of studies the authors have conducted from the development to the compression-moulding of anisotropic powders, and presents the results of on-circuit evaluation of anisotropic Nd-Fe-B bonded magnets on small motors.

2. Production Process

Fig. 2 shows the production process of anisotropic Nd-Fe-B bonded magnets from powder preparation to compression-moulding as studied by the authors. Flaky Nd-Fe-B ribbons were produced by the melt spinning process. The ribbons were vacuum packed in an iron container. The packing density of the ribbons can be raised to about 5 g/cm^3 by judiciously operating the press. The vacuum-packed ribbons are rolled at 700°C. This is the packed rolling process, the technology of which has been established to a maximum of 50 kg per batch on a commercial scale.

The as-rolled ribbons are bulky and must be ground again. Such a grinding method was selected that powder of good magnetic alignment could be produced as described later. The powder and thermosetting resin are then mixed to form a compound. The resin content of the compound ranges from 2 to 4 wt%. The compound is placed in between the die and punch, and is exposed to a magnetic field. Then, the easy direction of magnetization of magnet powder is aligned with the direction of the applied magnetic field, and the powder is compressed in the mould at a pressure of 4 to 8 tf/cm^2. The compact was cured in a temperature range from 100 to 120°C. The density of the bonded magnet is 5.8 to 6.2 kg/cm^3, which is about 80% of the true density of the Nd-Fe-B alloy. The bonded magnet is formed in the net shape and therefore does not need any further machining.

Detailed descriptions follow on the packed rolling and compression moulding processes.

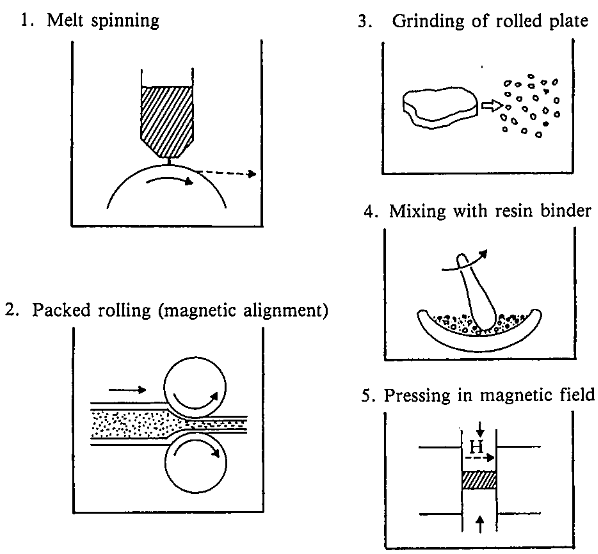


Fig. 2 Nd-Fe-B anisotropic bonded magnet production process

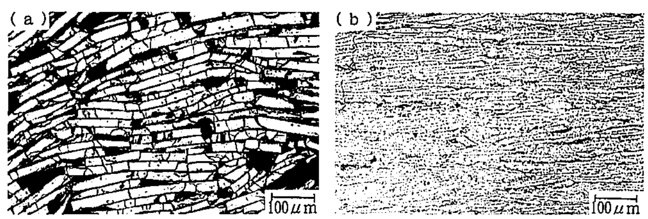
3. Crystallographic Alignment by Packed Rolling

(1) Alignment process

Nd-Fe-B ribbons prepared by melt spinning are composed of $Nd_2Fe_{14}B_1$ compound with 50 to 100 nm fine isotropic grains. With composition in the vicinity of $Nd_{14}Fe_{80}B_6$ (Nd-rich) in terms of atomic percentage, the powder of melt spun ribbon can be consolidated in a short time by hot pressing at 700°C. At this temperature, moreover, it is known to be crystallographically oriented through plastic deformation[2]. The $Nd_2Fe_{14}B_1$ compound is a tetragonal compound having easy magnetization axis which is easily magnetized in the c-axis direction. Plastic deformation, however, aligns the c-axis of individual grains parallel to the compressive stress direction. If this phenomenon occurs during the rolling process, an Nd-Fe-B plate rolled with the c-axis aligned parallel to the rolling reduction direction should be obtained.

As shown in **Photo 1**, the packed rolling process consolidates the melt-spun ribbons and reduces their thickness through plastic deformation. The authors measured the rolling reduction of the ribbons and minutely examined the corresponding changes in their density and magnetic alignment respectively for the subsurface and mid-thickness regions of the rolled material[7].

The results are as outlined in **Fig. 3**. As the rolling reduction advances, the powder density increases first in the subsurface region, followed by the mid-thickness region. When the rolling reduction rate exceeds 50%, the whole rolled bulk approximates the true density of an alloy. The residual magnetic flux density



(a) Before rolling (b) After rolling

Photo 1 Density increase and thickness decrease by rolling of rapidly solidified ribbons

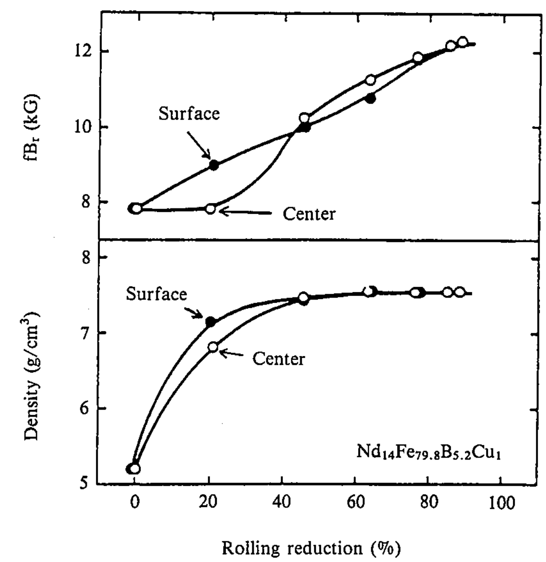


Fig. 3 Density and through-thickness residual magnetic flux density B_r on packed rolling

B_r was measured in respective regions to investigate the change in the degree of orientation of the c-axis due to the rolling reduction. **Fig. 3** shows the results. The residual magnetic flux density B_r rises first in the surface where the density is increased first. Rolling to 80% or higher reduction rate increased the B_r to 12 kG or more which corresponds to 80% or more in the degree of orientation, irrespective of the through-thickness position in the rolled material.

Fig. 4 shows the demagnetization curves of the Nd-Fe-B powder before rolling and after rolling by 88%. The final maximum energy $(BH)_{max}$ is 37.9 MGOe, which is about three times the initial value. This $(BH)_{max}$ value of 37.9 MGOe is comparable to that obtained by the conventional hot pressing, die upsetting (HD) process[8]. The present packed rolling process is characterized by ease with which large quantities can be processed compared with the HD process.

(2) Thermal stability of anisotropic powder

The rolled ribbons are bulky, but are easily cracked and therefore cannot be used as magnets in that state. They are milled again, bound by resin, and used as bonded magnets. The anisotropic powders described here refer to milled crystallographically aligned and rolled bulk ribbons.

Thermal stability has been a problem with anisotropic powders since the beginning of the development work. When a permanent magnet is once heated and then cooled, it generally cannot recover its original magnetic field strength. This is called irreversible magnetic flux loss. This tendency is great in the case of Nd-Fe-B anisotropic powders. The probable cause of the irreversible magnetic flux loss is grain coarsening during crystallographic alignment due to plastic deformation. The authors studied elements to be added to the Nd-Fe-B system to solve the problem and found that copper is effective for the purpose.

The irreversible magnetic flux loss decreases with increasing coercive force. The addition of copper improves the coercive force of the rolled ribbon plate. **Fig. 5** shows the coercive force $_iH_c$ after rolling and its change through heat treatment. Characteristically, the coercive force monotonically decreases with increasing annealing temperature for the copper-free ribbons, while it increases in the annealing temperature range from 500 to 700°C for the copper-bearing ribbons. From differential thermal scanning analysis (DTA), the authors found that a low-melting eutectic phase composed of CuNd and Nd is formed at grain boundaries, and concluded that the smoothing of the grain boundaries by melting the eutectic phase improves the coercive force[4].

Fig. 6 shows the irreversible magnetic flux loss of anisotropic bonded magnets. The irreversible magnetic flux loss of the copper-bearing magnets decreases in correspondence with the improvement of coercive force. The change in the coercive force through heat treatment is also reflected in the magnitude of the irreversible magnetic flux loss. The coercive force of copper-bearing magnets can be controlled by heat treatment, where heat resistance at 120 to 160°C can be assured. This high heat resistance allows

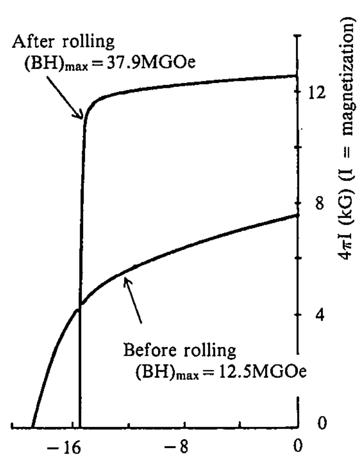


Fig. 4 Demagnetization curves before rolling and after 88% rolling

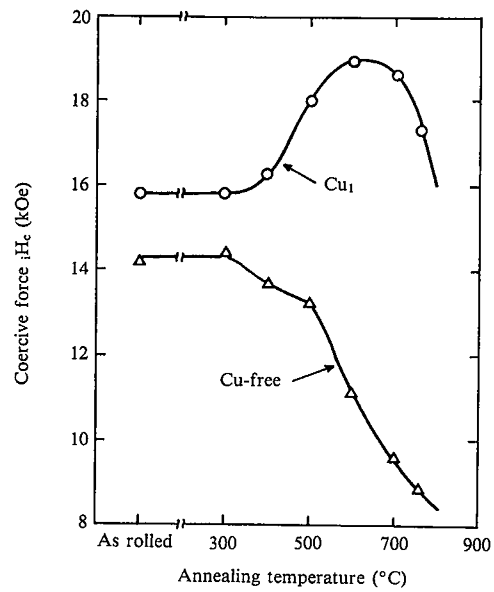


Fig. 5 Change in coercive force of rolled ribbons with annealing temperature
Cu-free: $Nd_{14}Fe_{80}B_6$
Cu_1: $Nd_{14}Fe_{79.8}B_{5.2}Cu_1$

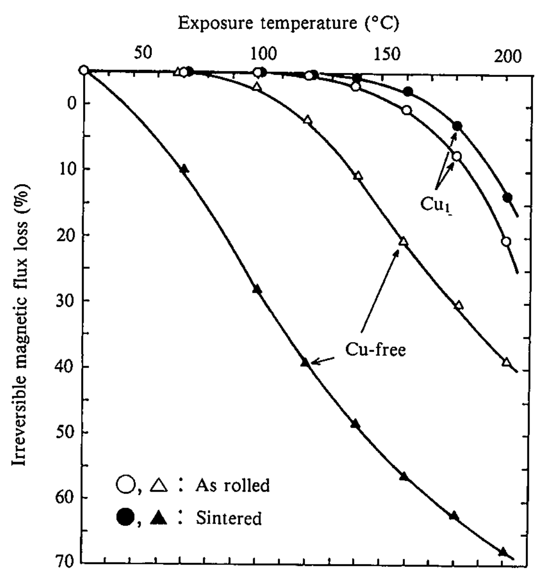
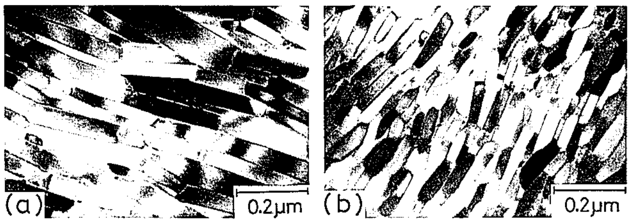


Fig. 6 Change in irreversible magnetic flux loss with temperature (permeance coefficient:2)



(a) $ND_{14}Fe_{80}B_6$ (b) $ND_{14}Fe_{79.8}B_{5.2}Cu_1$

Photo 2 Microstructures of rolled magnet materials

anisotropic bonded magnets to be used in motors for general office automation (OA) and factory automation (FA) equipment.

Photo 2 shows the microstructures of rolled ribbons as observed by transmission electron microscopy (TEM). The fine $Nd_2Fe_{14}B_1$ grains are flattened by the rolling operation. The size of the $Nd_2Fe_{14}B_1$ grains is markedly small for the copper-bearing ribbons and is 0.13 μm as average of maximum length of grains. This grain refinement is believed to make great contributions to the improvement of coercive force and the decrease in the temperature coefficient of coercive force, that is, a decrease in the irreversible magnetic flux loss. The temperature coefficient of coercive force is $-0.48\%/°C$ for 1% Cu-added ribbons and $-0.60\% °C$ for copper-free ribbons.

4. Pressing in Magnetic Field

The anisotropic powder is mixed with a resin binder and pressed in a magnetic field. As shown in **Fig. 7**, there are three methods of magnetic field pressing: perpendicular field pressing with the magnetic field direction H normal to the pressing direction P; parallel field pressing with the magnetic field direction H parallel to the pressing direction P; and radial field pressing with the magnetic field direction H normal and radiated to the pressing direction P. The magnetic field pressing method must be changed to suit the shape and property requirements of specific magnet parts. Ring-shaped or pipe-shaped magnets with powder particles magnetically aligned in the radial direction, or the so-called radially-oriented anisotropic bonded magnets, are in the largest demand for small-motor applications. Introduced below are some new findings obtained from the magnetic field pressing of Nd-Fe-B anisotropic bonded magnet powders:

(1) Effect of shape of powder particles

When a magnetic field is applied to magnet powders, individual magnet powder particles are rotated by the force of the magnetic field and are aligned to orient their c axis parallel to the direction of the magnetic field. The powder in that condition is pressed in the magnetic field to increase its density. In this process of densification, the magnetic alignment of the powder particles is destroyed to some extent, and the degree of destruction largely depends on the shape of the powder particles.

Two types of powders (particle size $< 297\ \mu$m) were prepared by different milling methods and were fabricated into bonded magnets by the perpendicular field pressing method. **Photo 3** shows the cross-sectional microstructures of the bonded magnets fabricated from the powders of two types, A and B. The c-axis direction of individual powder particles can be observed from the direction of etching-revealed ribbon boundaries (streaks in the particles) in the photo. The c-axis direction of each powder particle is indicated by an arrow in the lower part of each photo. The powder A has the directions of the arrows aligned better than the powder B. The residual magnetic flux density B_r, an index of magnetic alignment, is 9% higher for the bonded magnet from the powder A than for the one from the powder B. The powder A was pulverized by an impact grinding mill, while the powder B was pulverized by a shear grinding mill. The powder B is characterized by a large amount of flattened particles. Since the relationship between the magnetic field direction and the pressing direction for the radial field pressing was the same as for perpendicular field pressing, it was decided to use the powder A in the experiment described below.

(2) Aligning field dependence

The magnetic alignment of magnet powders varies with the strength of the magnetic field applied. **Fig. 8** shows the residual magnetic flux density B_r of the bonded magnets as a function of the magnetic field strength applied. The magnet powder par-

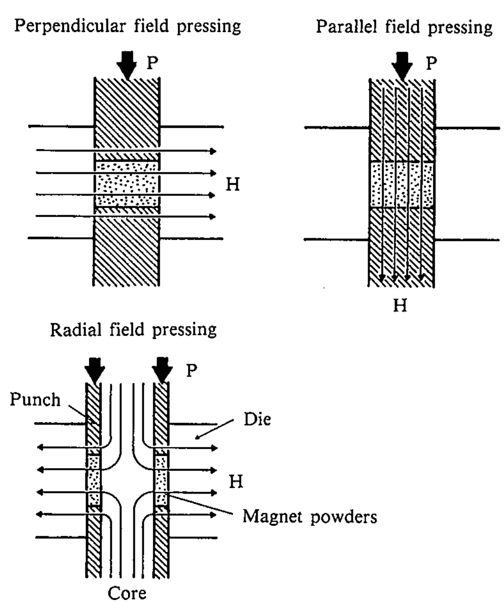


Fig. 7 Three methods of magnetic field pressing

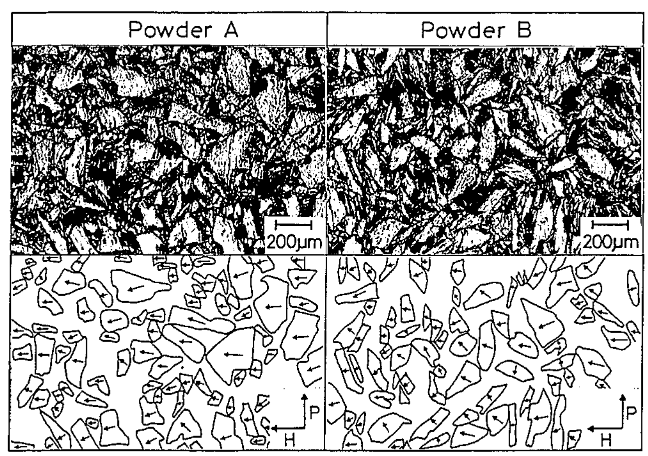


Photo 3 Cross-sectional microstructures of bonded magnets pressed in perpendicular magnetic field
Arrow for each particle indicates the axis of magnetization.

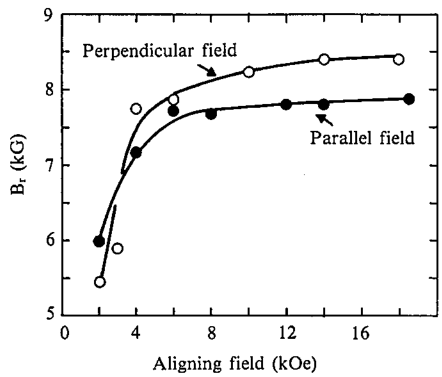


Fig. 8 Aligning field dependence of residual magnetic flux density B_r of bonded magnets

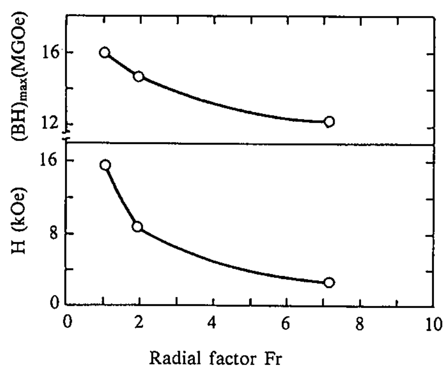


Fig. 9 Radial factor F_r versus magnetic field strength H and $(BH)_{max}$ of bonded magnets

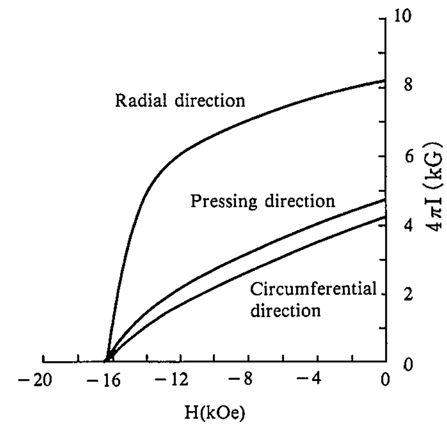


Fig. 10 Demagnetization curves of radially-oriented bonded magnets (F_r = 1.9)

ticles are aligned well by a relatively low magnetic field strength of about 2 to 4 kOe in both the perpendicular and parallel magnetic fields. This contrasts with the conventional Sm_2Co_{17} bonded magnets that require a strong magnetic field for magnetic alignment[5]. The fast rise in the initial magnetization of the Nd-Fe-B system anisotropic powders is responsible for its easy magnetic alignment in a relatively low magnetic field. This makes for advantage under the radial field pressing method described next.

The radial field is formed when the magnetic field passing through the core material is repulsed by the magnetic field coming from the opposite direction as shown in **Fig. 7**. Therefore, a strong magnetic field cannot be produced by a magnet with a small diameter and long length. The radial factor F_r can be conveniently introduced for representing the relationship between the magnetic field strength and the die size[9]. If a is the core diameter, b is the inside wall diameter of the die, and h is the height of the die, the radial factor F_r is given by

$$F_r = 2bh/a^2$$

The equation represents that the inside wall area of the die to be reached by the radial field is divided by the cross-sectional area of the core through which the repulsive field passes. Accordingly, F_r expresses the divergence degree of the radial field. As F_r increases from 1 to 2, the radial field is halved in strength.

Fig. 9 shows the magnetic field strength measured with three types of dies and the properties of the bonded magnets produced in the magnetic field in relation to the radial factor F_r. The maximum energy product $(BH)_{max}$ of the bonded magnets decreases with increasing F_r but attains to more than 15 MGOe when F_r is less than 2. **Fig. 10** shows the magnetic properties of the Nd-Fe-B radial anisotropic bonded magnets as measured in the radial, circumferential, and height (i. e., pressing) directions when F_r is 1.9. The magnetic properties measured in the radial direction show the highest values and indicate good magnetic alignment in the radial direction.

Conventional Sm_2Co_{17} bonded magnets with a small diameter and long length, were not fully magnetically aligned, and were sometimes unable to provide high magnetic properties. The application of Nd-Fe-B system anisotropic powders should alleviate these problems and make bonded magnets of high magnetic properties available over a wide size range.

5. Motor On-circuit Evaluation

Motors of smaller size and thickness are called for in the peripheral equipment of computers. Here are described the results of using the newly developed Nd-Fe-B bonded magnet on a spindle motor for 3.5-inch floppy disk drive (FDD). A prototype magnet (36 mm outside diameter × 32.5 mm inside diameter × 3 mm height) was made for a spindle motor of the construction shown in **Fig. 11**. It was evaluated in comparison with an isotropic Nd-Fe-B bonded magnet and an anisotropic Sm_2Co_{17} bonded magnets, both in current use.

Nd-Fe-B system anisotropic powders were prepared by the packed rolling process and were pressed in a radial magnetic field to produce an anisotropic bonded magnet with B_r = 8.4 kG, $_iH_c$ = 10.4 kOe, and $(BH)_{max}$ = 15.9 MGOe. The motor on-circuit evaluation results of the new bonded magnet are compared with those of the conventional bonded magnets, as shown in **Table 1**. The effective magnetic flux of the Nd-Fe-B anisotropic bond-

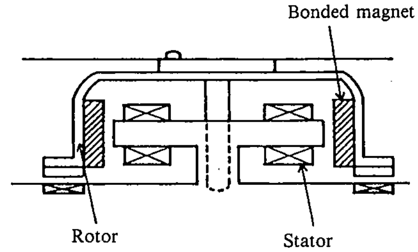


Fig. 11 Spindle motor used for on-circuit evaluation

Table 1 Results of on-circuit evaluation on spindle motor for FDD

Magnet	Effective magnetic flux (Mx)	Rated current (mA)
Nd-Fe-B anisotropic bonded magnet (newly developed)	689	155
Nd-Fe-B isotropic bonded magnet	570	171
Sm-Co anisotropic bonded magnet	618	163

ed magnet is 21% and 11% higher than that of the Nd-Fe-B isotropic bonded magnet and the Sm_2Co_{17} anisotropic bonded magnet, respectively. This means that the Nd-Fe-B anisotropic bonded magnet can be used to reduce the magnet height (the motor thickness) to provide the same motor output. The rated current of the Nd-Fe-B-Cu anisotropic bonded magnet is also lower than that of the conventional bonded magnets and is expected to contribute to the reduction of electricity consumption.

6. Conclusions

Nd-Fe-B anisotropic bonded magnets meet the needs of the times, but are not yet commercially applied. The reasons cited for this situation are: 1) concern about the thermal stability of anisotropic powders; 2) unavailability of low-cost production processes; and 3) unclarity of advantages of anisotropic bonded magnets. The authors solved the problem 1) by adding copper, proposed a packed rolling process for the problem 2), and made clear for the problem 3) that the Nd-Fe-B anisotropic powders can be magnetically aligned in a weak magnetic field and is thus suited for radial field pressing. Motor on-circuit evaluation demonstrated that the new bonded magnet can provide better magnetic properties than the conventional bonded magnets.

A completely new production process[10] that uses hydrogen is proposed for the Nd-Fe-B anisotropic powders, research in which field is predicted to become more active. The Nd-Fe-B anisotropic bonded magnets are not yet fully endorsed by good cost performance. They need further improvement in two important aspects functional enhancement and cost reduction achieved on the production materials side and development of new application fields on the product utilization side.

7. Acknowledgment

The authors express gratitude to Mr. Masayuki Ishikawa of Sankyo Seiki Mfg. Co., Ltd. for cooperation in the motor on-circuit evaluation of the Nd-Fe-B anisotropic bonded magnets.

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