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High-Energy NdFeB Magnets and Their Applications

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In sintered NdFeB magnets, additive elements for increasing coercivity decrease residual magnetization. Also, fine magnetic powder oxidization prevents identification of the stoichiometry composition ($Nd_2Fe_{14}B$). To improve the magnetic properties of a sintered NdFeB magnet, the authors have developed a method involving two alloys. Magnetic $Nd_2Fe_{14}B$ alloy and a rare-earth-rich alloy (including richer dysprosium content) are melted individually and mixed together after coarse pulverization. After the sintering process, dysprosium in the sintered body is enriched in each grain region near the grain boundary. The two-alloy method minimizes the liquid phase necessary to keep the coercive force at a useful level, and thus results in compositions closer to stoichiometry. The energy product of magnets having an inhomogeneous dysprosium distribution is typically 360 kJ · m^{-3} (45 MG·Oe) at production level. The corrosion characteristics of cobalt-substituted NdFeB magnets also were investigated. These magnets are now used in voice coil motors for hard disk drives and contribute to shortening access time and hard-drive downsizing.

Keywords

rare earth magnet, Nd2Fe14B, two-alloy method, dysprosium distribution, corrosion resistance, electroless nickel plating, voice coil motor

1. Introduction

THE energy product of sintered NdFeB magnets (hereafter called Nd magnets) has reached 418kJ · m^{-3} (52.3 MG · Oe) in the laboratory (Ref 1), whereas the $Nd_2Fe_{14}B$ stoichiometry compound (hereafter referred to as 2-14-1 compound) has a potential of 512 kJ · m^{-3} (64 MG·Oe) (Ref 2). Previously, the highest grade of sintered Nd magnets used in production had an energy product of 320 kJ · m^{-3} (40 MG · Oe) and insufficient coercivity. In order to obtain a higher residual magnetization (B_r), iron-rich compositions are necessary close to the 2-14-1 compound. However, this results in two problems: powder oxidation and a decrease in B_r. The fine powder of NdFeB alloys, with diameters of 3 to 4 µm, is easily oxidized in air to form Nd_2O_3. The oxygen weight in sintered Nd magnets is 0.5 to 0.7%. Because the neodymium element is consumed with oxidation and nonmagnetic Nd_2O_3 lowers the magnetic property, it is difficult to achieve an iron-rich composition close to the 2-14-1 compound at production level. A balance between oxidation protection and decreased use of additives is key to obtaining a high-performance Nd magnet. Moreover, since sintered Nd magnets have poor corrosion resistance, a coating is necessary for actual use. This paper describes a two-alloy method for improving the magnetic properties of Nd magnets. Corrosion resistance through the use of a cobalt-substituted NdFeB alloy is also discussed.

2. Powder Metallurgy Process

A two-alloy method for mass production (Ref 3) has been developed that differs slightly from the authors' previous work (Ref 4) and from that reported in Ref 1 and 5. Although the basic process employs the usual powder metallurgy method, two alloys are used. One alloy has a composition closely approximating the 2-14-1 compound, and the other has a composition rich in rare-earth elements and cobalt. The former is responsible for the intrinsic magnetic properties. The latter is a sintering aid and is responsible for the densification of the sintered body and the cleaning of sintered particle grain boundaries during the sintering process. In the rare-earth-rich alloy, more than half of the rare-earth content is dysprosium, and cobalt is heavily substituted. The two alloys are melted individually, then mixed after coarse pulverization. The remainder of the process

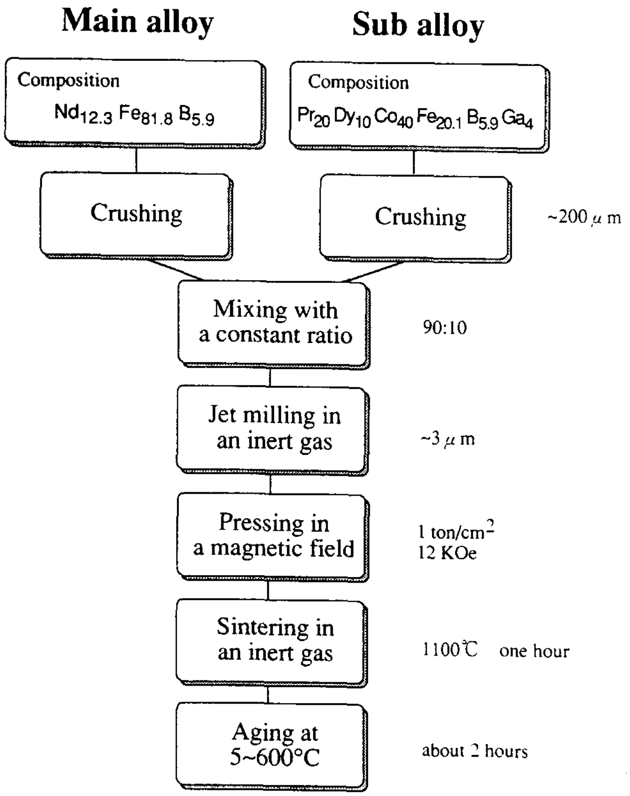

Fig. 1 Process flow of the two-alloy method

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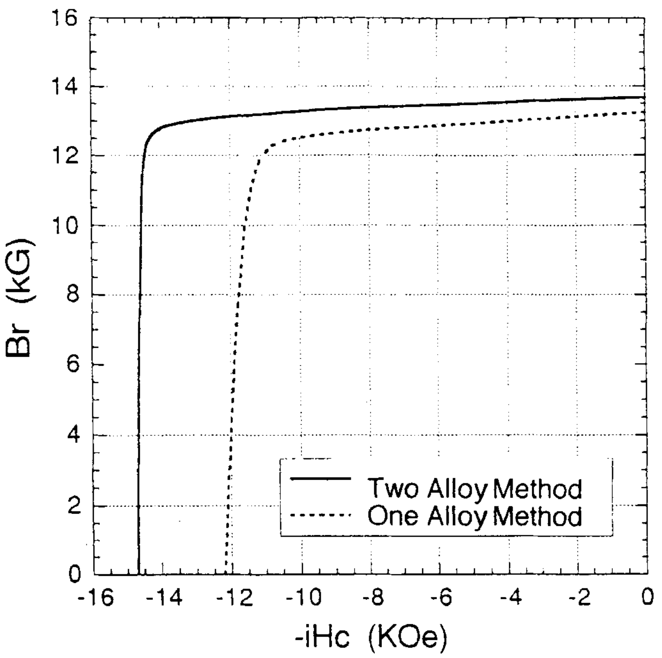


Fig. 2 Magnetization curve of sintered Nd magnets of the same composition produced by the two-alloy method and by the conventional method

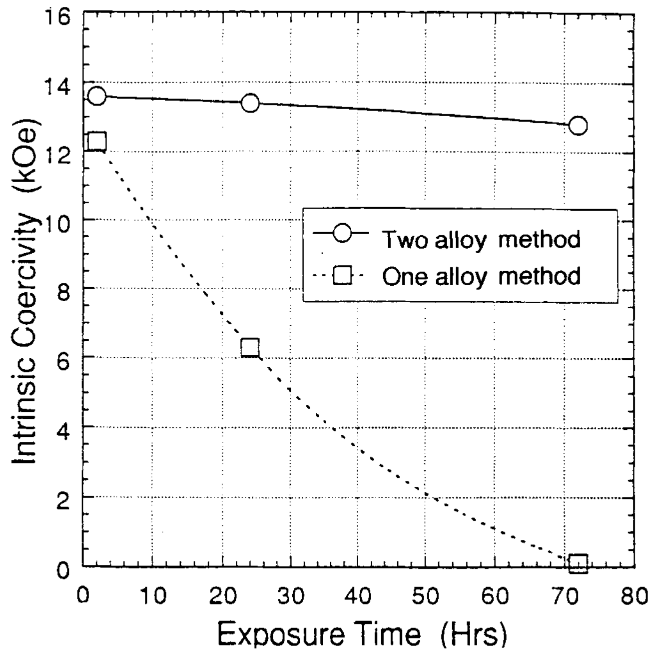


Fig. 3 Coercivity degradation of sintered magnets produced by the two-alloy method and the conventional method after exposing the fine powder in air at 25 °C and 40% relative humidity

Table 1 Composition of the as-cast rare-earth-rich alloy

Phase	Composition
1	$(Pr_{0.431}Dy_{0.569})_{17.7}(Fe_{0.379}Co_{0.621})B_{13.21}Ga_{1.23}$
2	$(Pr_{0.418}Dy_{0.582})_{24.2}(Fe_{0.383}Co_{0.617})B_{0.35}Ga_{2.18}$
3	$(Pr_{0.644}Dy_{0.356})_{31.9}(Fe_{0.295}Co_{0.705})B_{3.72}Ga_{2.00}$
4	$(Pr_{0.984}Dy_{0.016})_{60.4}(Fe_{0.046}Co_{0.954})B_{2.82}Ga_{1.23}$

is identical to the usual method shown in Fig. 1. As is characteristic of this process, pressing fine powder in a homogeneous magnetic field is possible to accomplish in air.

In the two-alloy method, many possibilities exist as to which combination is most suitable. The authors have examined many rare-earth-rich alloy compositions and alloy mixing ratios, and have determined an optimum combination. The main alloy possesses the stoichiometry alloy composition $Nd_{12.3}Fe_{81.8}B_{5.9}$. Because a large amount of primary iron exists in the as-cast alloy, solid-solution heat treatment is necessary to obtain the single 2-14-1 phase. The composition of the rare-earth-rich alloy is $Pr_{20}Dy_{10}Co_{40}Fe_{20.1}B_{5.9}Ga_{4}$. The cobalt-rich addition improves the corrosion resistance of the fine powder and the sintered body. The additions of dysprosium, praseodymium, and gallium enhance coercivity. The mixing ratio is approximately 90:10. After sintering, the resulting composition is $Nd_{11.3}Pr_{1.6}Dy_{0.8}Fe_{76.9}Co_{3.1}B_{5.9}Ga_{0.3}$.

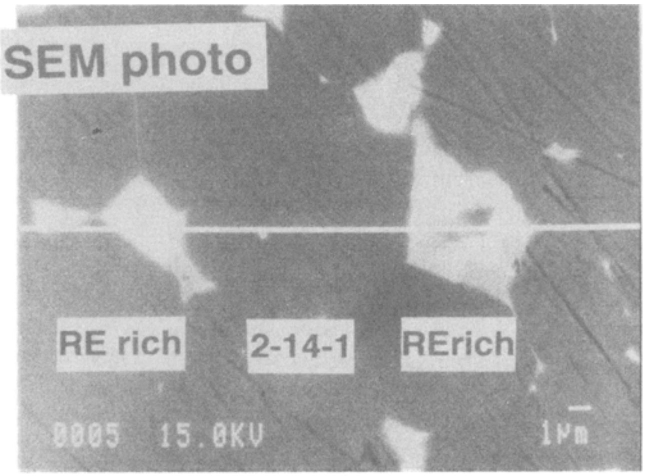
3. Magnetic Properties of Nd Magnets Produced by the Two-Alloy Method

Figure 2 shows the magnetic properties of Nd magnets of the same final composition produced by the two-alloy method and by the conventional method. The magnet produced by the two-alloy method is superior.

Figure 3 shows the oxidation stability of the fine powder of both types of magnets exposed to air at 24 °C and 40% relative humidity. The coercive force of the sintered magnet produced conventionally decreases rapidly when left in the air for 1 day. However, the coercive force of the sintered magnet produced by the two-alloy method remains almost constant after 3 days, a great improvement in powder stability.

The low oxidation level of the fine powder is attributed to the rare-earth-rich alloy, which has a high cobalt content. Usually, the rare-earth-rich alloy powder is more prone to oxidation than the 2-14-1 powder. The as-cast rare-earth-rich alloy is composed of four phases (Table 1). Each phase has been identified by electron probe microanalysis and x-ray diffraction and has a cobalt-rich composition. Cobalt-base compounds have good corrosion resistance, so powder oxidation in air is substantially suppressed. The two-alloy method thus allows the fine powder to be handled in air during the pressing process, making special treatments such as vacuum or inert gas unnecessary. Therefore, this method is suitable for mass production. The two-alloy method also reduces the oxygen content in the sintered body and improves its corrosion resistance. The cobalt addition to the 2-14-1 alloy further suppresses oxidation.

The magnetic property improvement is attributed primarily to the dysprosium distribution and partially to the praseodymium distribution. Dysprosium initially exists only in the rare-earth-rich alloy, and then diffuses to the 2-14-1 powder during the sintering process. The diffused dysprosium is distributed mainly near the grain boundaries of the sintered 2-14-1 particles (Fig. 4). Although dysprosium near grain boundaries increases coercivity, it decreases saturation magnetization. However, the absence of dysprosium in the center region re-



	Place	Nd	Dy	Fe	Co	B	Ga
1	RE rich	77.2	3.87	2.1	0.15	0.15	0.02
2	2-14-1	25.8	2.62	67.8	2.89	0.91	0.26
3	2-14-1	26.8	1.77	68.0	2.84	0.82	0.26
4	2-14-1	27.8	0.40	68.5	2.92	0.98	0.29
5	2-14-1	27.8	0.18	68.5	2.83	1.02	0.28
6	2-14-1	27.8	1.38	66.6	2.88	0.91	0.20
7	RE rich	76.6	3.55	4.1	0.20	0.18	0.04

Fig. 4 Element distributions in a Nd magnet produced by the two-alloy method

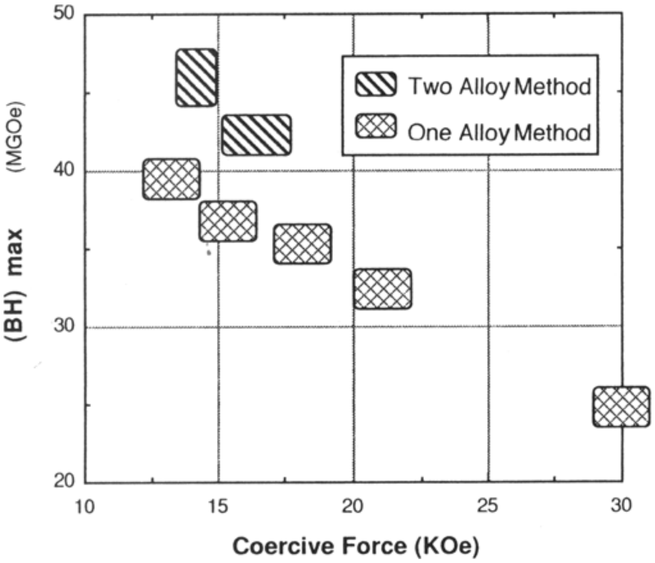


Fig. 5 New Nd magnet series produced by the two-alloy method

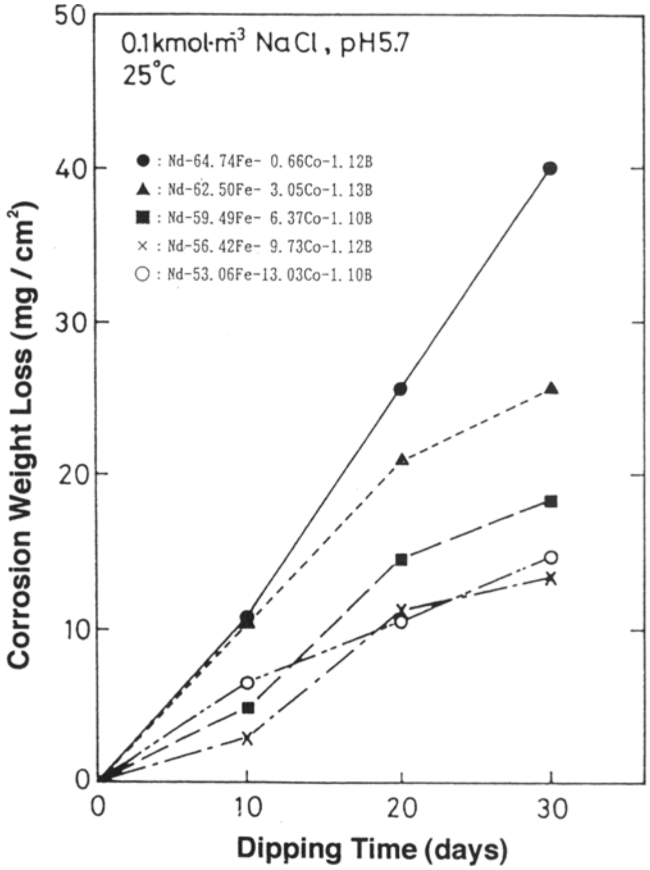


Fig. 6 Corrosion weight loss of NdFeCo(B) magnets against dipping time in 0.1 kmol · m^{-3} NaCl solution

duces this decrease in magnetization, thus yielding a high-performance Nd magnet.

Coercivity and saturation magnetization form a complementary relationship when the additives are introduced. If both the ratio of praseodymium and dysprosium in the rare-earth-rich alloy and the mixing ratio of the two alloys are varied, the superior Nd magnet series shown in Fig. 5 can be obtained. Also, when the fine powder used in the two-alloy method is treated in an inert gas or in a vacuum, an even higher magnetic property can result.

4. Corrosion Resistance of NdFeB Alloys

As previously mentioned, the addition of cobalt improves the fine powder oxidation of NdFeB alloys as well as the sintered magnet corrosion resistance. These phenomena have been reported by many researchers (Ref 6). Moreover, it has been reported that multiadditives, including cobalt, have further improved sintered magnet corrosion resistance (Ref 7). However, there are a few papers that detail the corrosion characteristics of cobalt-substituted Nd magnets (Ref 8).

The corrosion weight loss of NdFeCo(B) magnets against cobalt content is shown in Fig. 6. The alloys were dipped into 0.1 kmol · m^{-3} NaCl neutral solution at pH 5.7 and 25 °C for 24 h. Corrosion weight loss decreases rapidly when the cobalt content is increased. The potentiostatic anode polarization curves of the same sintered magnets are shown in Fig. 7. Its condition was a sweep speed of 23 mV/min in 0.1 kmol · m^{-3} NaCl solution of pH 5.7 at 25 °C. The passivation state was not apparent despite the cobalt addition, but it was noted that the corrosion potential becomes noble as the cobalt content increases. Although the corrosion resistance of a cobalt-substituted Nd magnet is improved, it is not sufficient without the coating. The poor corrosion resistance of sintered NdFeB magnets is attributed to a local cell formation in the rare-earth-rich phase. Therefore, the rare-earth-rich constituents need further investigation with regard to the potentiostatic anode polarization analysis.

In the two-alloy method, the rare-earth-rich alloy constituents shown in Table 1 are only slightly different from those of the conventional method. However, these compounds were not found in the sintered magnet. It is assumed that they reacted during sintering and changed to the 2-14-1 phase and the conventional rare-earth-rich phase. The rare-earth-rich phase in the sintered magnet is composed of a cobalt-base rare-earth compound and a gallium-rich compound. Detailed analysis is a subject for further research.

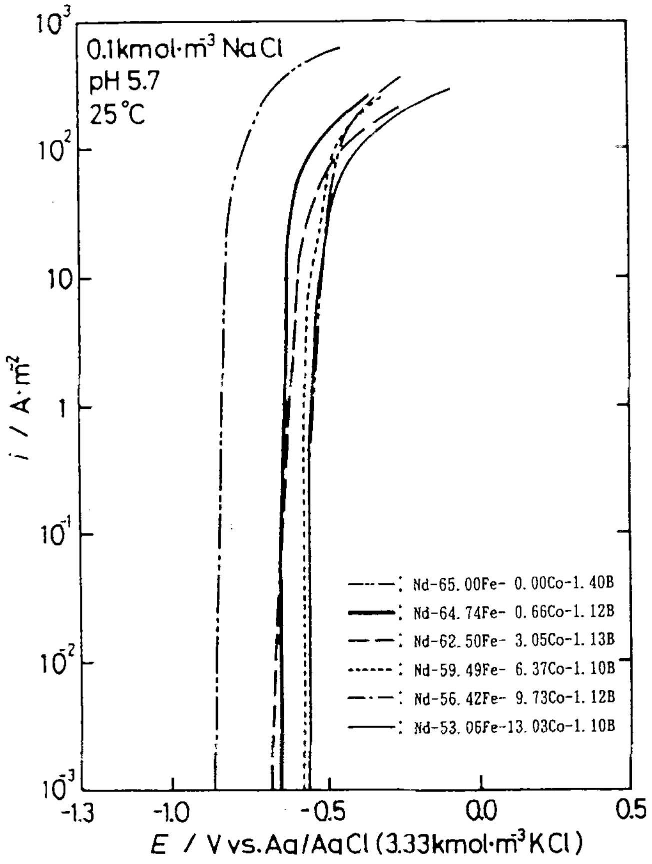


Fig. 7 Potentiostatic anode polarization curve of NdFeCo(B) magnets in 0.1 kmol · m^{-3} NaCl solution of pH 5.7 at 25 °C

The sintered magnet produced by the two-alloy method has the same corrosion resistance as that produced conventionally. A coating of some kind is necessary in the sintered Nd magnet for corrosion protection. An electronickel plating developed by the authors' group is the most popular coating (Ref 9). Although the coating has excellent corrosion resistance and good adhesive strength, a few problems exist. Film thickness at the magnet edges is two or three times thicker than at the magnet center because of the edge effect of electric current. This inhomogeneity is a serious problem for a narrow-gap circuit using thin magnets, such as a small-size voice coil motor (VCM). In order to improve the film thickness distribution, electroless nickel plating has been developed. It is composed of a copper underlayer and a nickel upper layer; total thickness is about 10 µm. Because the adhesive force between the electroless nickel layer and the Nd magnet is weak, the electroless copper layer, which has good contact with the Nd magnet, is necessary. The pretreatment process involved in electroless plating is almost the same as that used in electroplating. The film thickness of the electroless nickel plating is almost constant at any position, including the edge. Also, it retains the same level of corrosion resistance and adhesive force as the electronickel plating.

5. Voice Coil Motor Application

Many flat-coil-type VCMs are used in small-size hard disk drives for head access. Use of a high-performance Nd magnet produced by the two-alloy method allowed a higher gap flux to be obtained. For comparison of the conventional magnet (Shin-Etsu's N36) and the new magnet (Shin-Etsu's N45), Fig. 8 shows the magnetic flux and the torque constant in a flat-coil VCM gap. The torque constant achieved by the new magnet is 10% greater than that achieved by the conventional magnet, thus resulting in higher acceleration velocity.

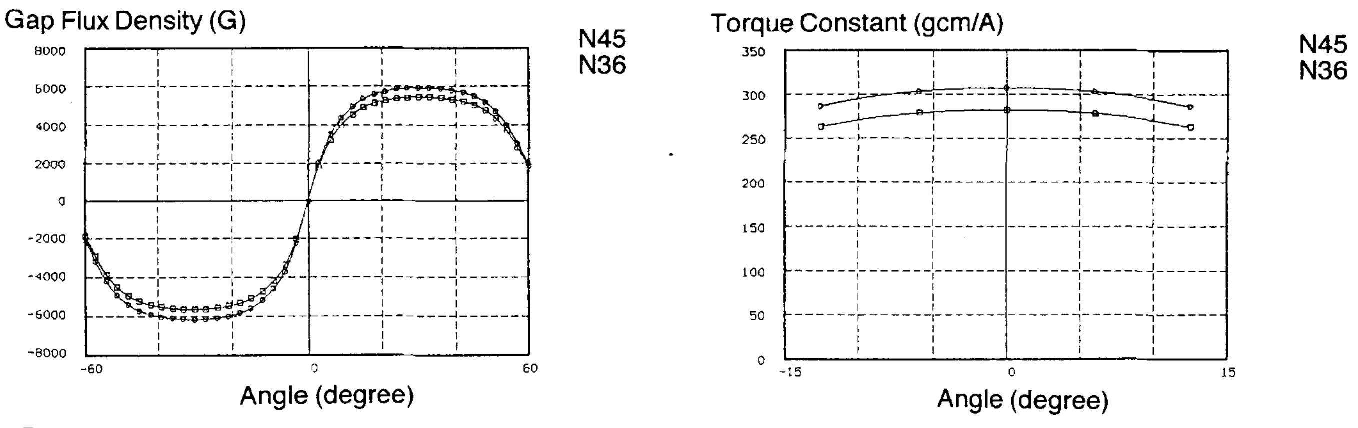


Fig. 8 Magnetic flux distribution and torque constant distribution for an N45 magnet (two-alloy method) and an N36 magnet (conventional method)

6. Conclusions

- A new method of preparation involving two alloys has been developed to produce magnets with compatible coercivity and residual magnetization properties.
- Sintered Nd magnets with energy products of 360 kJ · m^{-3} (45 MG · Oe) and moderate coercivity have been mass produced using the two-alloy method.
- Fine powder oxidation and corrosion resistance of the sintered magnet body were improved by cobalt-rich substitution in the rare-earth-rich alloy. Although the cobalt substitution improved corrosion resistance, it was not sufficient.
- In order to improve the homogeneity of the coating film thickness, an electroless nickel plating process has been developed.
- The high-performance Nd magnet produced by the two-alloy method is used in VCM applications.

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