ELSEVIER

Journal of Alloys and Compounds 281 (1998) 64–68

Journal of ALLOYS AND COMPOUNDS

Improvements of irreversible thermal losses in anisotropic $Nd_2Fe_{14}B$-based resin-bonded permanent magnets made from HDDR powder

T. Ikegami, M. Uehara, S. Hirosawa*

Fundamental Research Section, R&D Division, Sumitomo Special Metals Co., Ltd., 2-15-17 Egawa, Shimamoto-cho, Mishima-gun, Osaka 618, Japan

Abstract

The relationship between the intrinsic coercivity (H_{cJ}) and irreversible magnetic flux losses at elevated temperatures has been studied on compression-molded, resin-bonded anisotropic magnets made from $Nd_2Fe_{14}B$-type hard magnetic powders prepared by means of the hydrogenation–disproportionation–desorption–recombination (HDDR) process. Powders with various H_{cJ} values were prepared by partial substitution of Dy for Nd, and the temperature dependence of H_{cJ} of these powders were investigated. The H_{cJ} values range from 0.86 to 1.3 MA m^{-1} at room temperature. A rule of thumb has been found that the minimum room temperature value of H_{cJ} required to reduce the irreversible losses to the level comparable to the values encountered in rapidly-solidified, isotropic $Nd_2Fe_{14}B$ (H_{cJ}=0.76 MA m^{-1}) magnets is 1.2 MA m^{-1}. The difference in the H_{cJ} values to yield similar losses in these two types of materials is attributed to the difference in the temperature dependence of H_{cJ}. The dependence of the magnetizability on temperature of these magnets has also been studied. It is demonstrated that moderate heating of the magnets significantly increases the magnetic flux density obtainable below the magnetizing temperature when the magnetizing force is not large enough to saturate the magnet. In such cases, frequently encountered in actual applications, the magnetic flux density is stabilized in the temperature range below the magnetizing temperature.

Keywords: HDDR; Thermal losses

1. Introduction

The production and usage of rare earth bonded magnets have shown a remarkable increase, particularly since the invention of rapidly solidified $Nd_2Fe_{14}B$-type hard magnetic powders in 1983 [1]. These types of magnets are magnetically isotropic. Although there are many advantages of being isotropic [2], they cannot exploit the full potential of the hard magnetic phase. The average energy product of the isotropic $Nd_2Fe_{14}B$-type resin-bonded magnets is typically 80 kJ m^{-3}, which is far smaller than the theoretical limit of approximately 326 kJ m^{-3} for an ideal 80% loaded anisotropic magnet.

Anisotropic $Nd_2Fe_{14}B$ hard magnetic powder was first obtained by crushing hot-deformed $Nd_2Fe_{14}B$ bulk magnets [3], but commercial utilization of this kind of powder appears to have been unsuccessful. More recently, an innovative process called hydrogenation–disproportionation–desorption–recombination (HDDR) has been developed to produce anisotropic $Nd_2Fe_{14}B$-type hard magnetic powder [4]. The maximum energy product of 170 kJ m^{-3} was achieved on compression-molded resin-bonded magnets using this type of powder [5]. The anisotropic HDDR-processed $Nd_2Fe_{14}B$-type powder is now available commercially, and it is expected to penetrate into application fields where resin-bonded magnets with magnetic performance superior to conventional isotropic Nd–Fe–B are desired.

One of the major concerns about anisotropic resin-bonded magnets based on the $Nd_2Fe_{14}B$ phase has been the stability of the magnetic properties at elevated temperatures. As a consequence of the relatively large temperature dependence of the intrinsic coercivity (H_{cJ}), which amounts approximately to −0.55%/K, a large irreversible magnetic flux loss results, especially when the magnet is heated. With typical H_{cJ} values in the 0.9–1.0 MA m^{-1} range, the highest operating temperature of the $Nd_2Fe_{14}B$-type anisotropic bonded magnets is about 60°C.

In order to expand the usable temperature range, the present authors have recently developed high-coercivity magnetic powders of the $Nd_2Fe_{14}B$-type with H_{cJ} values exceeding 1.2 MA m^{-1} [6]. The high coercivity was achieved by utilizing the HDDR process in conjunction with alloy modification, that is, a partial substitution of Dy

*Corresponding author. E-mail: hirosawa.s@ssmc.co.jp

0925-8388/98/$ – see front matter
PII: S0925-8388(98)00773-7

for Nd. However, improvement of the thermal stability along this direction creates another difficulty, namely, a larger coercivity should lead to a larger magnetizing force required to saturate the magnet. This can be a major obstacle in certain applications in which small, multi-pole magnets are used, because the magnetizing force available in magnetizing fixtures confined to small spaces may be limited. The purpose of this paper is to propose a practical way of effectively overcoming this problem in order to use the high-coercivity anisotropic hard magnetic powder in resin-bonded magnets.

One of the possible ways of solving this problem is to raise the temperature of the magnet during the magnetizing procedure. Adoption of this method should also lead to stabilization of the magnetic properties with respect to the irreversible thermal losses in a temperature range lower than the magnetizing temperature. In this paper, the magnetic properties of the Dy-containing anisotropic hard magnetic powder and their temperature dependence are briefly reported, and the relation between the room temperature H_{cJ} values and irreversible flux losses is studied in order to clarify the minimal H_{cJ} level required for practical applications. In addition, the magnetizability at elevated temperatures is studied. It is demonstrated that it is beneficial to magnetize the high-coercivity $Nd_2Fe_{14}B$-type HDDR-processed magnets at elevated temperature in that both higher magnetic flux density and reduced irreversible thermal losses can be achieved in comparison to those obtainable by room temperature magnetization.

2. Experimental procedures

Several classes of anisotropic hard magnetic powder with various H_{cJ} values were prepared from alloys of chemical compositions $(Nd_{1-x}Dy_x)_{12.8}Fe_{65.6}Co_{15}Ga_{0.5}Zr_{0.1}B_6$. The alloys were prepared by induction melting and casting into a metal mold. After homogenizing the cast alloys in an Ar atmosphere at 1120°C for 16 h, they were crushed and subject to the HDDR treatment in an atmosphere-controlled furnace. In the HDDR treatment, the alloys were subject to a hydrogenation process under a 0.15–0.20 MPa hydrogen atmosphere in a range of temperature between 850 and 890°C for 3 h and a subsequent desorption process under vacuum in the same temperature range. After swift cooling, the powder was taken out of the furnace and loosened by sieving, and then used for preparation of resin-bonded magnets. The magnetic properties of the powder were measured using a vibrating sample magnetometer (VSM) on samples classified into 63–106 μm particle size.

Bonded magnets were prepared by molding a mixture of the HDDR-treated powder and 3 wt.% epoxy resin at 100°C under a magnetic field of 0.8 MA m^{-1} applied perpendicular to the pressing direction. The maximum pressing pressure was 0.7 GPa. The molded compacts were cured at 150°C. For comparison, magnets were prepared from commercial isotropic powder produced by means of rapid solidification (MQP-B) under identical molding and curing conditions.

The open magnetic flux of the sample magnets was measured using a conventional flux meter in order to evaluate irreversible flux losses and magnetizability of the magnets. Irreversible flux losses were measured on magnets of a permeance coefficient of approximately 2 which were magnetized by a pulsed magnetic field with a maximum height of 5 MA m^{-1} and a rise time of approximately 1 ms. The magnetizability, that is, the dependence of the open magnetic flux on the external magnetizing force, was measured on magnets with a permeance coefficient of approximately 3 which were initialized by demagnetizing with a direct current (dc) magnetic field after preparation.

3. Results and discussion

3.1. Magnetic properties of the HDDR powder and their bonded magnets

Examples of the magnetic properties at room temperature of $(Nd_{1-x}Dy_x)_{12.8}Fe_{65.6}Co_{15}Ga_{0.5}Zr_{0.1}B_6$ hard magnetic powders are shown in Fig. 1 as a function of Dy content. In this figure, only results obtained for the following set of HDDR processing parameters are shown: hydrogen pressure 0.2 MPa, hydrogenation temperature 875°C, and desorption temperature 850°C. A relatively steep increase in H_{cJ} for $x<0.025$ is evident. For larger Dy content, the increment in H_{cJ} becomes small at the further expense of the magnetic flux density.

The temperature dependence of H_{cJ} of resin-bonded magnets is shown in Fig. 2 for $x=0$ and 0.025. For comparison, the temperature dependence of H_{cJ} of an

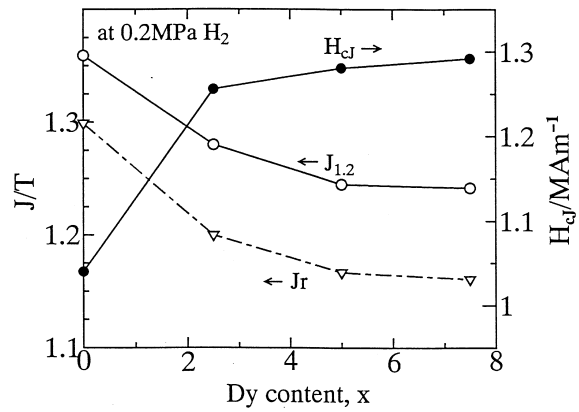

Fig. 1. Dependence of room temperature magnetic properties on Dy content in anisotropic $(Nd_{1-x}Dy_x)_{12.8}Fe_{65.6}Co_{15}Ga_{0.5}Zr_{0.1}B_6$ powder produced by the HDDR process. $J_{1.2}$ stands for magnetic polarization under an applied magnetic field of 1.2 T.

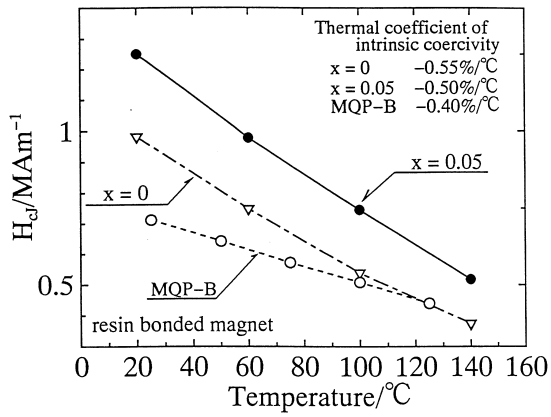

Fig. 2. Temperature dependence of the intrinsic coercivity of anisotropic $(Nd_{1-x}Dy_x)_{12.8}Fe_{65.6}Co_{15}Ga_{0.5}Zr_{0.1}B_6$ powder for $x=0$ and 0.025.

isotropic bonded magnet made from rapidly solidified $Nd_2Fe_{14}B$-type powder (MQP-B) is also shown. The temperature coefficient of H_{cJ} in the temperature range between 20 and 140°C is −0.55%/K for $x=0$ and −0.50%/K for $x=0.025$. These values are to be compared to the −0.40%/K for the isotropic magnet.

3.2. Irreversible flux losses

The relationship between irreversible flux losses and room temperature values of H_{cJ} is shown in Fig. 3. The irreversible flux losses in this study are defined as the relative change of open magnetic flux at room temperature from the initial saturation-magnetized state to a state after the magnet was exposed to a certain temperature (100°C maximum) in air for 1 h. Reflecting the difference in the temperature coefficient of H_{cJ}, a magnet prepared from

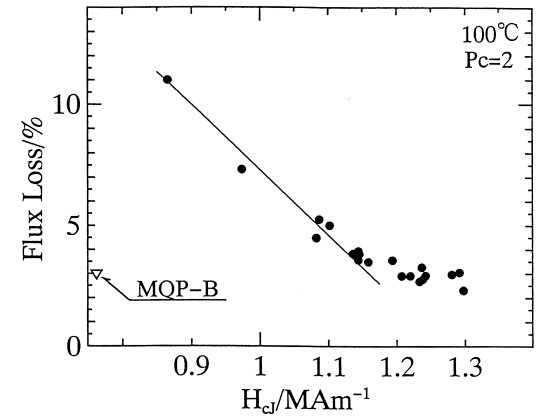

Fig. 3. Relationship between irreversible flux losses of resin-bonded magnets of a permeance coefficient of 3 made from anisotropic $Nd_2Fe_{14}B$-type powder and their intrinsic coercivity at room temperature. The irreversible losses were measured after exposure of the magnets to 100°C ambient atmosphere for 1 h. Specimens before the test recorded magnetic flux values (in 10^5 Maxwell Turn as observed as the output values of a magnetic flux meter) of 1.89 for the one made from HDDR powder of $H_{cJ}=1.24$ MA m^{-1} and 1.50 for the one made from the MQP-B powder.

anisotropic powder with a H_{cJ} value of 0.9 MA m^{-1} shows an irreversible loss of nearly 10%, whereas the isotropic, rapidly-solidified magnet shows only about 3% loss. The irreversible flux loss of the anisotropic magnets decreases with increase of the room temperature H_{cJ} value. In order to reduce the flux losses to the level of conventional MQP-B (approximately 3%, see Fig. 3), a room temperature coercivity of approximately 1.2 MA m^{-1} is required for the anisotropic magnets. According to Fig. 3, the improvement in irreversible thermal loss is insignificant for H_{cJ} values greater than 1.2 MA m^{-1}. This is a consequence of the degradation of the shape of the demagnetization curves which occurs for Dy content >0.025.

3.3. Magnetizability at elevated temperatures

The dependence of the relative magnetic flux on magnetizing force in dc-demagnetized anisotropic resin-bonded magnets with H_{cJ} values of 1.0 and 1.24 MA m^{-1} is shown in Fig. 4 for samples with a permeance coefficient of 3. The onset of magnetization is shifted by approximately the same amount as the increment in the H_{cJ} value. The suppressed rise of the relative open flux is a consequence of the magnetic history of the hard magnetic powder in the course of fabrication. Namely, the powder has experienced the alignment magnetic field and been magnetized. Dc demagnetization does not bring the magnet back to the thermally demagnetized state [7].

The effect of magnet temperature in the magnetizing procedure is shown in Fig. 5. In this figure, the magnetizability of the high-coercivity magnet with a H_{cJ} value of 1.24 MA m^{-1} under relatively low magnetizing forces is compared for three temperatures (20, 60, and 80°C) of magnets being magnetized. The relative open flux is measured at 20°C and is normalized against the open flux obtained with the magnet magnetized by a 5 MA m^{-1}

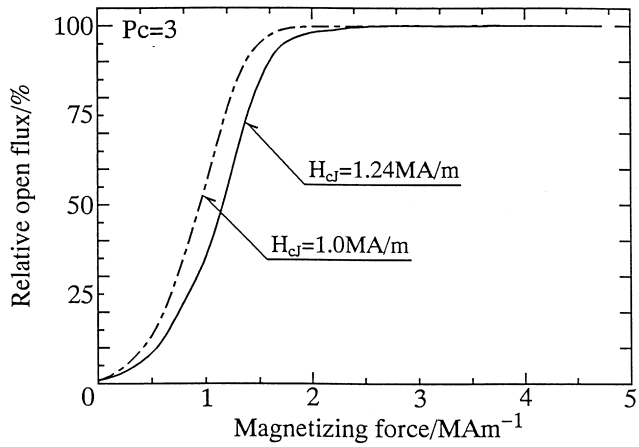

Fig. 4. Dependence of the relative magnetic flux on magnetizing force for resin-bonded magnets made from anisotropic $Nd_2Fe_{14}B$-type powders of intrinsic coercivity 1.0 and 1.24 MA m^{-1}. The temperature of the magnets during the magnetizing procedure is room temperature.

Table 1
Room temperature magnetic properties of anisotropic resin-bonded magnets magnetized at 20 and 60°C under a relatively small magnetizing force of 1.2 MA/m. The magnets were made from anisotropic hard magnetic $(Nd_{0.975}Dy_{0.025})_{12.8}Fe_{65.6}Co_{15}Ga_{0.5}Zr_{0.1}B_6$ powder prepared by the HDDR process

Magnetizing temperature (°C)	B_r (T)	H_{cJ} (MA m^{-1})	$(BH)_{max}$ (kJ m^{-3})
20	0.763	0.944	104
60	0.845	0.963	129

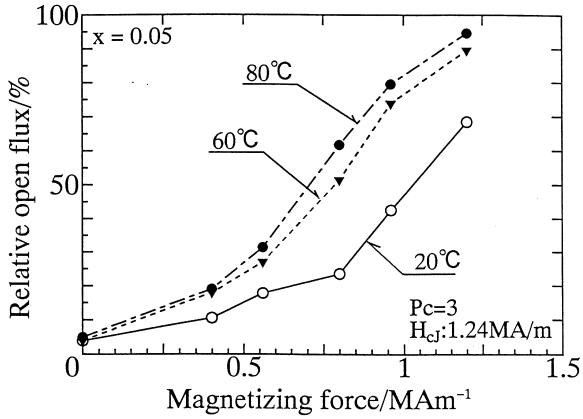

Fig. 5. Effect of magnet temperature during the magnetizing procedure on magnetizability of the magnets. Dependence of room-temperature relative magnetic flux on magnetizing force is shown for three magnetizing temperatures, 20, 60 and 80°C, for resin-bonded magnets made from anisotropic $Nd_2Fe_{14}B$-type powder with intrinsic coercivity of 1.24 MA m^{-1}.

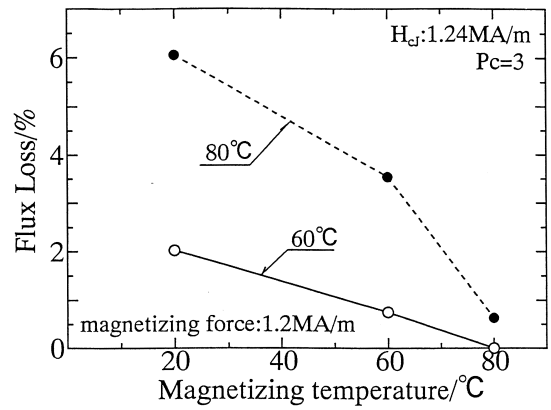

Fig. 6. Dependence of relative irreversible flux losses on magnetizing temperature for a resin-bonded magnet made from anisotropic $Nd_2Fe_{14}B$-type powder magnetized under a relatively small magnetizing force of 1.2 MA m^{-1}. The intrinsic coercivity of the hard magnetic powder is 1.24 MA m^{-1} and the permeance coefficient of the magnet is 3.

pulse field at 20°C. A significant increase in the room temperature value of the open magnetic flux is obtained by modestly heating the magnets during the magnetizing procedure. Indeed, nearly 95% saturation is achieved by a magnetizing force of 1.2 MA m^{-1} when the magnet is heated to 80°C. The room temperature magnetic properties of unsaturated magnets magnetized under a 1.2 MA m^{-1} field at 20 and 60°C are compared in Table 1. The magnets were made from anisotropic hard magnetic $(Nd_{0.975}Dy_{0.025})_{12.8}Fe_{65.6}Co_{15}Ga_{0.5}Zr_{0.1}B_6$ powder, which shows a saturated H_{cJ} value of 1.24 MA m^{-1}.

These results indicate the importance of controlling the temperature of the magnet in actual manufacturing of magnetic devices using this type of magnet. Namely, if the temperature of the magnets changes from time to time, constant magnetic performance cannot be obtained when the available magnetizing force is insufficient to saturate the magnet, which is frequently the case encountered in confined magnetizing fixtures for small, multi-pole magnets.

3.4. Irreversible losses of the unsaturated magnets

The dependence of the magnetic flux losses on the temperature at which magnets are magnetized under a magnetizing force which is insufficient to saturate them is shown in Fig. 6 for anisotropic bonded magnets of H_{cJ}= 1.24 MA m^{-1}. The magnitude of the magnetizing force is 1.2 MA m^{-1}, which is typical in practical magnetizing fixtures. The irreversible losses decrease with increasing temperature of the magnets in the magnetizing procedure (indicated in the figure). When magnetized at 80°C, the magnet shows no irreversible loss for the 60°C exposure. For the 80°C exposure, the loss is well below 1%.

The present results demonstrate the importance and effectiveness of controlling the temperature of magnets above room temperature during the magnetizing procedure for anisotropic resin-bonded magnets made from $Nd_2Fe_{14}B$-type magnetic powders, especially when the available magnetizing force is limited. Another application would be the post-assembly magnetizing method, that is, performing the magnetizing procedure after assembly of magnetic devices. This will greatly reduce the difficulty in handling magnetized powerful permanent magnets. The moderate heating technique in the magnetizing procedure would be indispensable in some applications in order to fully exploit the potential magnetic performance of anisotropic resin-bonded Nd–Fe–B magnets.

References

[1] J.J. Croat, J.F. Herbst, R.W. Lee, F.E. Pinkerton, J. Appl. Phys. 55 (1984) 2083.

[2] J.J. Croat, J. Appl. Phys. 81 (1997) 4804.
[3] R.W. Lee, E.G. Brewer, N.A. Brandley, IEEE Trans. Magn. 26 (1985) 1958.
[4] T. Takeshita, R. Nakayama, in: Proceedings of the 10th International Workshop on Rare-Earth Magnets and Their Applications, Kyoto, Japan, 1989, Vol. 1, p. 551 (unpublished).
[5] S. Hirosawa, M. Uehara, S. Mino, N. Ishigaki, T. Tomida, J. Appl. Phys. 81 (1997) 4821.
[6] T. Ikegami, S. Hirosawa, in: Proceedings of the 9th International Symposium on Magnetic Anisotropy and Coercivity in Rare-Earth Transition Metal Alloys, Sao Paulo, Brazil, 1996, World Scientific, Singapore, 1996, p. 288.
[7] M. Uehara, T. Tomida, H. Tomizawa, S. Hirosawa, Y. Maehara, J. Magn. Magn. Mater. 159 (1996) L304.