Magnequench Magnets Status Overview

V. Panchanathan

The advent of neodymium-iron-boron materials having excellent magnetic properties and potential economic advantages has initiated a new era in permanent magnet technology. One method of making these magnets is by the rapid solidification process. It is typically carried out by melt spinning, which produces a highly stable, magnetically hard microstructure powder, directly from the melt. This can be used for bonded magnet applications. Alternatively, this powder can be hot pressed to produce fully dense isotropic magnets with energy products up to 15 MGOe. Anisotropic magnets with energy products ranging up to 50 MGOe can be produced by thermomechanical orientation or hot deformation process. Current processing and properties of Magnequench (General Motors) materials are reviewed, as well as the applications and advances of these materials. The advances include high-temperature bonded magnet and high-energy product anisotropic bonded and fully dense magnets.

Keywords
permanent magnets, Nd-Fe-B magnets, rapid solidification, bonded magnets, hot pressed magnets, hot deformed magnets, neodymium

1. Introduction

MAGNEQUENCH permanent magnets differ from other grades of Nd-Fe-B materials in that their manufacture involves a rapid solidification, or melt-spinning step, in which a stream of molten alloy is quenched to form a fine-grained, microstructurally stable powder. This powder can be used for both bonded magnets and fully dense intermetallic magnets with high energy products (Ref 1, 2, 3). The family of products marketed by Magnequench includes the isotropic powder itself (MQP), a resin-bonded magnet made from this material (MQ1), a hot-pressed magnet (MQ2), and the hot-deformed anisotropic material (MQ3). Energy products span a wide range from 5 MGOe for injection molded powders to about 50 MGOe for the MQ3 material. Recently, Magnequench introduced anisotropic powder made by the hydrogenation disproportionation desorption recombination (HDDR) process. This publication briefly reviews the current status and advances in Magnequench magnets.

2. Isotropic Bonded Magnets

Of all the Nd-Fe-B materials currently being marketed, the isotropic powder made by rapid solidification is unique because of its extremely fine microstructure. It is the only Nd-Fe-B material capable of being processed into bonded magnets. The grades of isotropic powders in production are given in Table 1. Powders C and D are similar to A and B, respectively, but with significantly improved temperature coefficient of residual induction, (∝), providing for greater stability in high-temperature applications. Powders B and D have higher residual induction combined with low intrinsic coercive force, H_{ci}, and are aimed at applications in which magnetization may be difficult. Further, MQP-D has a much improved knee value, which is due to the existence of a more uniform microstructure across the thickness of the ribbon. It is attributed to the higher amount of cobalt on the solidification rate and growth kinetics of the $Nd_2(Fe,Co)_{14}B$ grains.

Table 2 lists the properties of MQ1 bonded magnets produced from these powders. Manufacture includes blending the powder with epoxy, compaction molding to a density of roughly 6.1 g/cm^3, and then curing the epoxy at 170 °C for 30 min. Energy products achieved range from a nominal of 9

V. Panchanathan, Magnequench, Delphi-Energy & Engine Management Systems, U.S.A., Anderson, IN 46013, USA.

Table 1 Characteristics of isotropic powders

Type	Residual induction B_r, kG	Intrinsic coercive force H_{ci}, kOe	Temperature coefficient, ∝ %/°C at 25-100 °C
MQP-A	7.60	15.0	–0.130
MQP-B	8.20	9.0	–0.105
MQP-C	7.55	16.0	–0.070
MQP-D	8.00	10.5	–0.070
MQP-N	8.00	9.5	–0.130
MQP-O	7.45	12.5	–0.130

Table 2 Characteristics of bonded isotropic magnets

Type	Residual induction B_r, kG	Intrinsic coercive force H_{ci}, kOe	Maximum energy product $(BH)_{max}$, MGOe	Temperature coefficient, ∝ %/°C at 25-100 °C
High coercivity				
MQ1-A	6.5	15.0	9.0	–0.130
MQ1-C	6.5	16.0	9.0	–0.070
Medium coercivity				
MQ1-O	6.5	12.5	9.0	–0.130
Low coercivity				
MQ1-B	6.9	9.0	10.0	–0.105
MQ1-D	6.9	10.5	10.5	–0.070
MQ1-N	6.8	9.5	9.5	–0.130

Note: The units reported herein are cgs units. Conversion to SI units are as follows: Tesla = kG/10, kA/m = 79.5 × kOe, and kJ/m^3 = 7.95 × MGOe.

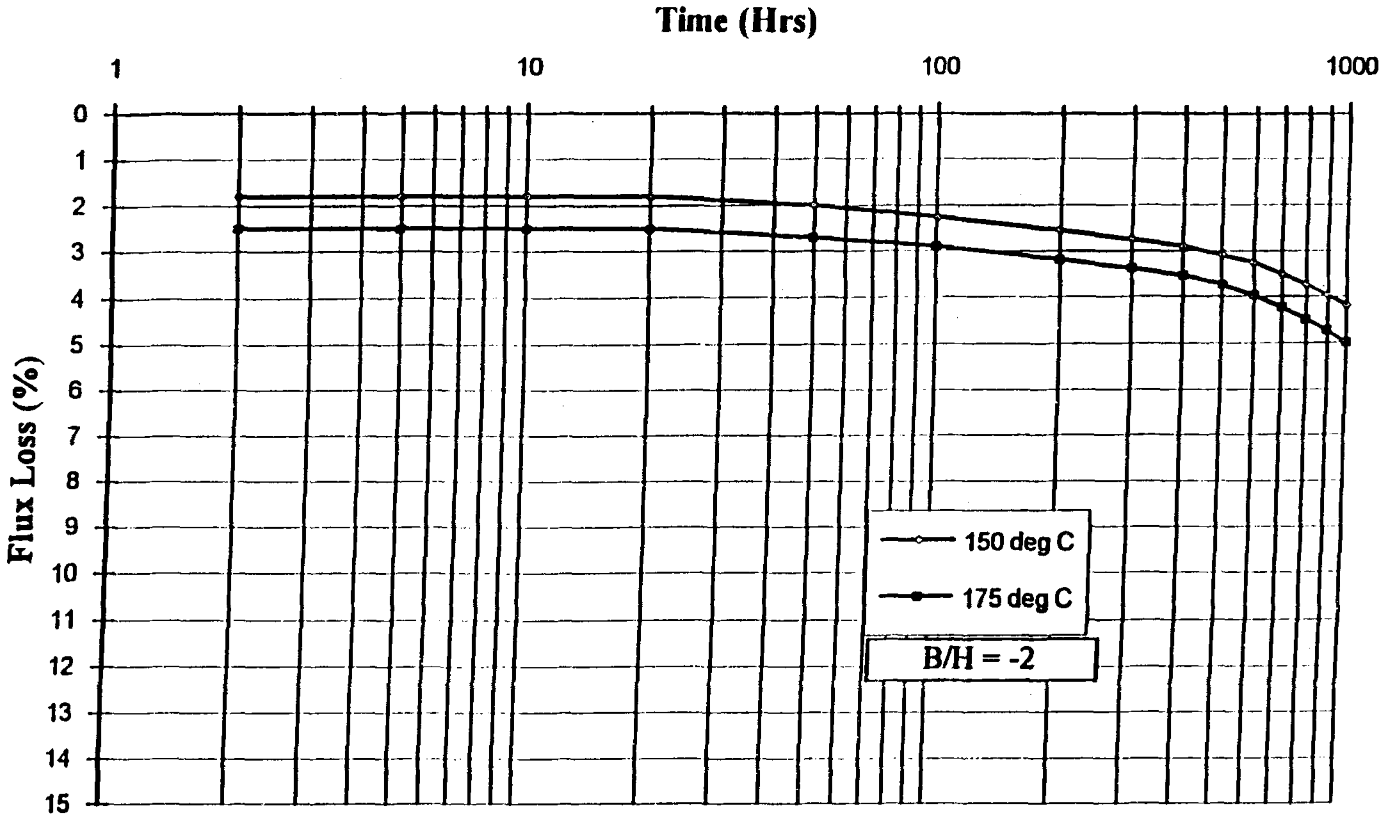

Fig. 1 Flux loss variation with time for bonded magnets

MGOe for the high coercivity powder to 10.5 MGOe for low coercivity powder (Table 2).

Magnequench recently introduced a new powder, MQP-N. It is essentially a substitute for MQP-B without any cobalt addition. With wide fluctuations in cobalt price, this powder should find applications for use at lower temperatures (<100 °C) than MQP-B. The characteristics of these powders are listed in Table 1.

The injection molded magnets using the isotropic powders are routinely used for temperatures above 125 °C; however, the compression molded magnets are normally restricted to temperatures below 125 °C. The joint development work between Magnequench and Daido Steel Company Ltd., Japan, resulted in the introduction of MQP-O. The magnetic properties of this powder are listed in Table 2. This powder contains 2.0 to 2.5 wt% niobium as an additive. While Magnequench rates this magnet as suitable for use at 140 °C using their epoxy binder, Daido Steel rates these bonded magnets for use at 175 °C using their proprietary binders (Ref 4) (Fig. 1). The total loss is reduced from 15 to 5% with this powder of which 6.5% is attributed to the niobium addition and 3.5% to the binder. This indicates that both the alloy composition and binder are important in the development of heat-resistant bonded magnets. The B/H in Fig. 1 refers to the load line of the magnet, which is a shape dependent value.

In recent years, the development activities in the isotropic powders have been concerned with improvements in B_r. Yamamoto et al. (Ref 5) reported a maximum energy product of 19.2 MGOe for vanadium-containing compositions. A bonded magnet made from this material had an energy product of 11.7 MGOe. Yajima et al. (Ref 6) reported B_r values of over 9 kG and energy products between 17 to 19 MGOe in zirconium-containing compositions. The recent studies on Nd-Fe-B magnets include a mixture of two different phases (a hard and a soft phase) with the aim of enhancing magnetic properties while reducing the overall Nd content compared to the standard $Nd_2Fe_{14}B$ magnet. These are also referred to as exchange coupled magnets. The microstructure consists of $Nd_2Fe_{14}B$ with predominantly either Fe_3B or αFe. European (Ref 7) and Japanese (Ref 8) researchers reported a B_r value of 12 kG and a H_{ci} value of 3.8 kOe in a Nd-Fe-B magnet having $Nd_2Fe_{14}B$ and Fe_3B phases. The microstructure and magnetic properties of a bonded and fully dense magnet produced from melt-spun ribbons of the composition $RE_{4.5}TM_{76}Ga_1B_{18.5}$ were reported by Mishra and Panchanathan (Ref 9). According to them, the demagnetization characteristics of the material are determined by the Fe_3B phase giving a remanence close to 0.79 of saturation value; also the moderate value of coercivity is due to the influence of the $Nd_2Fe_{14}B$ phase. High remanences and energy products also were reported in melt-spun ribbons with a fine-grained mixture of αFe and $Nd_2Fe_{14}B$ (Ref 10, 11). The gas atomization technique is also investigated for the production of Nd-Fe-B powders; however, at this time, all the production quantities are manufactured by the melt-spinning process.

Bonded neodymium magnets are now used in a wide range of applications. A number of reasons account for this growth.

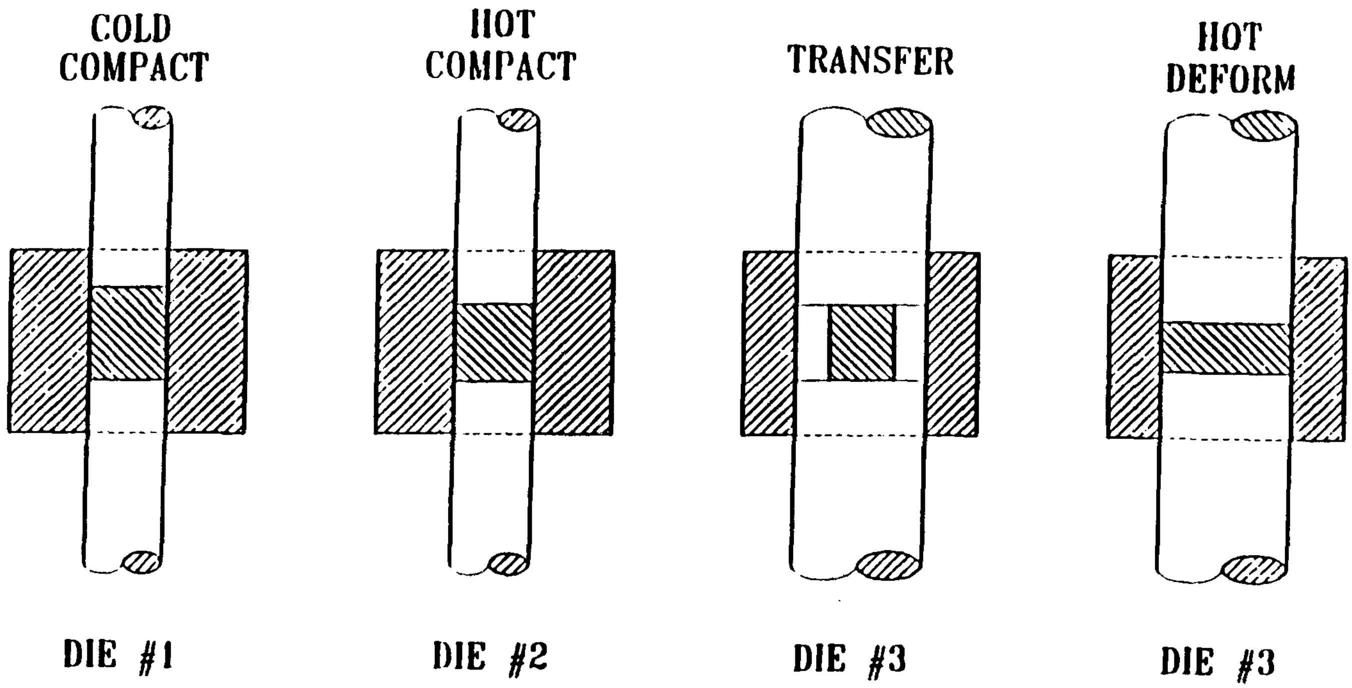

Fig. 2 Hot pressing, hot deformation process for making MQ2 and MQ3 magnets

First, the powder is highly stable and can be handled in air for long periods of time (Ref 1, 2). In addition, since the powder is isotropic, no alignment field is needed during bonding, which requires less expensive tooling and allows for much faster cycle times. The finished magnets can be magnetized in any direction, which can provide greater design flexibility. The green compacts before curing can be handled in manufacture with ease, and complex shapes can be produced easily with a high degree of precision. These bonded magnets have a substantially lower coefficient of intrinsic coercivity, approximately –0.4%/°C. This, in effect, means that for high-temperature applications, the lower B_r of the isotropic materials is partially offset. These isotropic materials are not magnetized during production; hence, there is less tendency for stray magnetic particles to adhere to the parts during coating and final component assembly. This is a significant advantage in the computer industry where such particles can seriously damage the operation of the memory.

One of the predominant applications of bonded neodymium magnets is in micromotors of various types. They are primarily for office automation, computer peripheral, and consumer electronic applications. Specific applications include printers, fax machines, video cameras, floppy disk drive actuators, and spindle motors for hard disk drives. Other applications include brush-type motors for cordless power tools and brushless motors for robots and home applications. Some automotive applications include instrument gauges, actuators, sensors, fuel pumps, and cranking motors. The development of bonded heat-resistant magnets should spur growth in automobile applications and other high-temperature applications.

A portion of the growth in the bonded MQ1-type market has been for new applications or for the replacement of existing bonded or sintered Sm-Co. Close to 50% of the growth has gone to replace sintered ferrite magnets. The primary motivation for this transition from ferrite to a comparatively more expensive Nd-Fe-B product is that it allows the designer to reduce the size and weight or alternatively increase the performance of the application. Future growth areas of MQ1-type magnets are automotive, appliances, and power tools.

3. Hot-Pressed Magnets

The fully dense magnets are obtained by hot pressing rapidly quenched ribbons (Ref 3). Above 725 °C, the material is relatively plastic, and consolidation can be rapidly achieved at relatively low pressures (50 to 60 MPa). The resultant magnet is isotropic and called MQ2 magnet. The process is shown in Fig. 2. In this method, a cold preform is made in die No. 1, lubricated, and then hot pressed in an open chamber under an argon stream in die No. 2. This open air hot pressing under an argon stream has simplified the whole operation and also improved the productivity. Table 3 lists the properties of MQ2-E and MQ2-F magnets, which are now in production. The energy product of both types of magnets is 14 MGOe. Figure 3 shows the irreversible losses with temperature for both types of magnets. Note the losses are less than 3% at 175 °C using MQ2-F. What is significant is that parts aged at 150 to 175 °C for 10,000 h show virtually no structural or permanent loss in magnetic properties (Fig. 4). These magnets have a substantially lower coefficient of intrinsic coercivity, approximately –0.46%/°C versus –0.65%/°C, for anisotropic materials at 100 °C. In fact, at higher temperatures, the values are still lower as shown in Fig. 5. In addition, the process lends itself to a continuous high volume operation, and since the part is hot pressed in a die, the resulting piece is near net shape requiring little finish grinding.

The current applications include sensors and cranking motors for automobiles (Ref 12). A good example is anti-lock

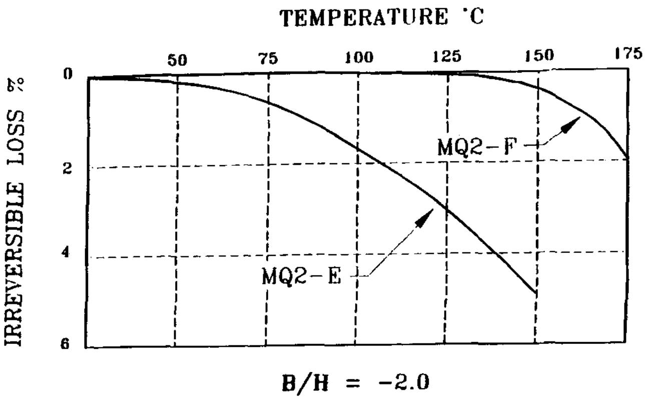

Fig. 3 Variation of irreversible losses with temperature for different MQ2 magnets

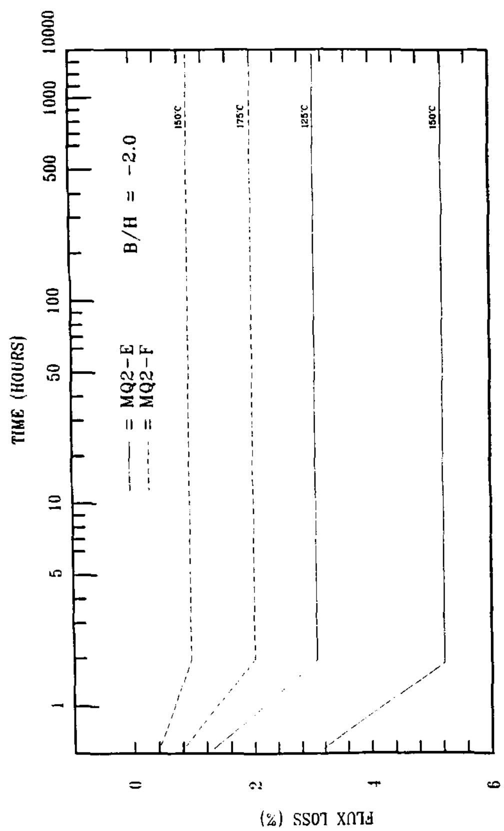

Fig. 4 Variation of flux loss with temperature for different MQ2 magnets

brakes, which have become widely available in the last few years. The need to downsize these systems, while at the same time improving response time, is driving this application toward higher performance, lower inertia motors, which will in turn require high-performance magnets. The other trends in automotive design include increased fuel economy, which will require reduce weight and higher efficiency motors and generators. Other applications include industrial motors and marine flywheel alternators. The growth areas for these magnets are with sensors and brushless motors with ring-shaped magnets for automotive and nonautomotive applications.

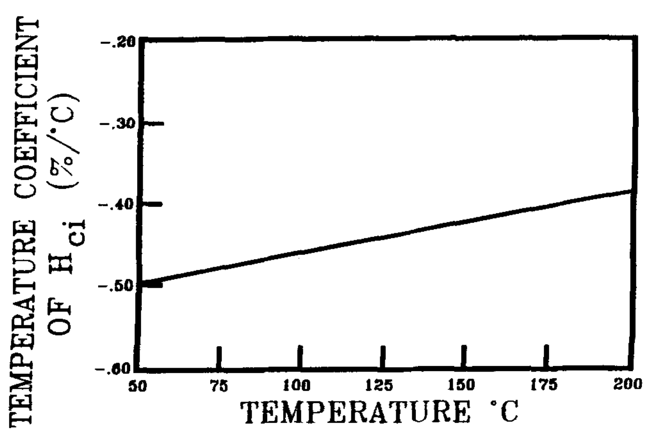

Fig. 5 Temperature coefficient of H_{ci} at different temperatures for MQ2 magnets

Table 3 Characteristics of hot worked magnets

Type	Residual induction (B_r), kG	Intrinsic coercive force (H_{ci}), kOe	Maximum energy product ($(BH)_{max}$), MGOe	Temperature coefficient, ∝ %/°C at 25-100 °C
Hot pressed—MQ2 (isotropic)				
MQ2-E	8.0	18.0	14.0	–0.10
MQ2-F	8.0	20.0	14.0	–0.09
Hot deformed—MQ3 (anisotropic)				
MQ3-E38	12.8	12.6	38	–0.10
MQ3-E43	14.0	12.0	45	–0.10
MQ3-F38	12.7	16.6	38	–0.09
MQ3-F43	13.6	15.7	43	–0.09
MQ3-G32	11.6	21.0	32	–0.09

4. Hot-Deformed Magnets

The manufacturing method is shown schematically in Fig. 2. The hot-pressed part to full density is made in an open chamber under an argon stream as detailed earlier. It is then transferred to die No. 3 and subjected to deformation or die upsetting operation. The magnet material flows outward to fill the die with the reduction in height of 50 to 70%. Processing occurs quite rapidly and at low pressures especially at temperatures above its plastic temperature of around 725 °C. Once again, this deformation is carried out in an open chamber, which has considerably simplified the operation leading to improved productivity. The fully dense magnet obtained is called MQ3 and is anisotropic. The alignment mechanism, which has been studied extensively by Mishra (Ref 13) is purely crystallographic, and no alignment field is applied. Table 3 lists the properties of MQ3-E, MQ3-F, and MQ3-G magnets, which are in production. The energy product ranges from about 32 to 43 MGOe, and the intrinsic coercivity ranges from 12 to 21 kOe. Figure 6 shows the variation in irreversible loss with temperature for the three types of magnets. Note the low irreversible losses at 175 °C using MQ3-G. This magnet is developed in response to customer's demand to have low irreversible losses at higher tem-

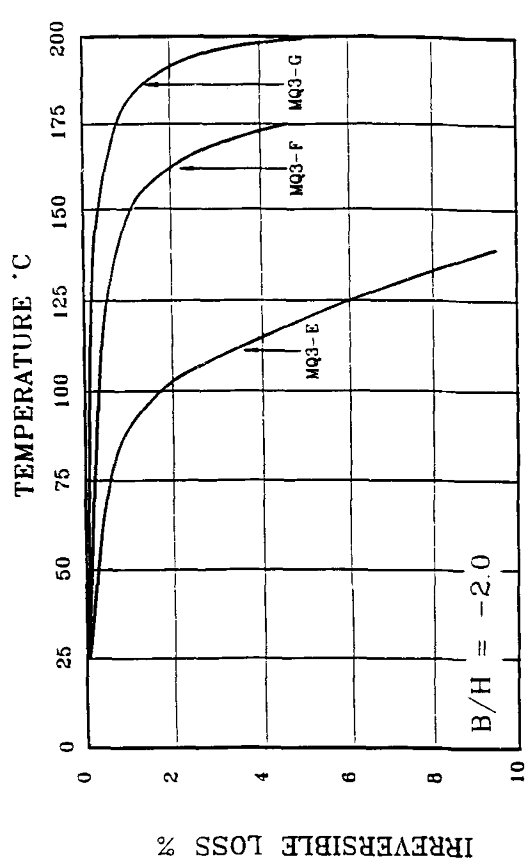

Fig. 6 Variation of irreversible losses with temperature for different MQ3 magnets

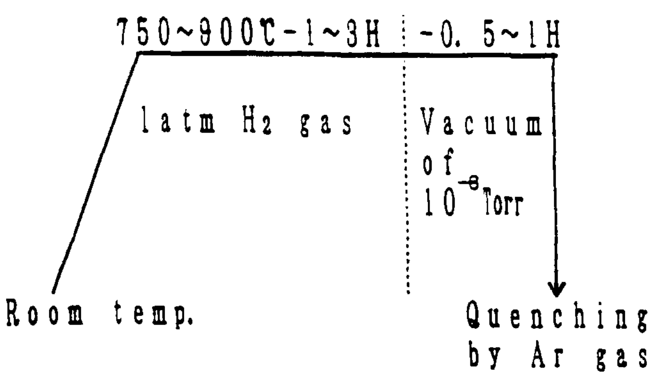

Fig. 7 Schematic diagram of HDDR process

peratures. MQ3 magnets have inherent corrosion resistance and excellent aging characteristics similar to hot-pressed magnets. Magnets aged between 150 and 175 °C for 10,000 h show virtually no structural loss in magnetic properties. Higher coercivities than those of hot-pressed magnets are needed for hot-deformed magnets to have lower irreversible losses. It is due to the higher value of temperature coefficient of intrinsic coercivity value of –0.65%/°C for MQ3 magnets compared to –0.46%/°C for MQ2 magnets.

The process lends itself to a continuous high volume operation, and since the part is formed in a die, the resulting piece is near net shape, requiring little finish grinding. These magnets can be processed readily to achieve radial alignment. Further, these magnets have excellent coatability and good magnet uniformity.

A major aim of developmental activity in recent years has been to improve the alignment or B_r of these hot-deformed materials. While the theoretical energy product of anisotropic Nd-Fe-B material ranges as high as 64 MgGOe, the highest values currently in production are roughly 43 to 45 MOe. The work has focused on several areas including the rapid solidification process itself. Another area to study is the effect of various additives on the magnetic properties and hot workability of hot deformed materials. The magnetic alignment is a result of crystallographic alignment during high-temperature deformation (Ref 13). Since remanence is directly related to the degree of alignment of the grains inside the material, processing and alloying variations that increased the fraction of aligned $Nd_2Fe_{14}B$ grains raise the remanence. High coercivity can be achieved by maintaining the fine grain sizes and dispersing an effective intergranular phase as a barrier to domain wall motion. By combining chemical and processing modifications, Mishra et al. (Ref 14) achieved an energy product of 48 MGOe in hot-deformed magnet. The interior of this magnet consisted of well-aligned $Nd_2Fe_{14}B$ grains separated by a Nd-rich intergranular phase. Intermixed with such aligned grains were zones of unaligned fine-grained material without any intergranular phase The hot-pressed material before deformation was found to contain $Nd_2Fe_{14}B$ (~50 nm) grains separated by a rare-earth-rich intergranular phase without large grains or inclusions. This intergranular phase is a key factor for deformation and alignment, and further enhancement in alignment is possible by controlling the redistribution of the intergranular phase during hot deformation.

Additives introduced in small quantities can have a substantial impact on the properties since they affect either the grain boundary phase or the $Nd_2Fe_{14}B$ phase. The effect of various additives on the hot workability and properties was reported earlier (Ref 2).

The effect of cobalt addition on magnetic properties of hot-deformed magnets was studied (Ref 15). Addition of 2.8 at.% (2.5 wt%) cobalt considerably improved the temperature coefficient of H_{ci} compared to the base alloy; however, further additions of cobalt did not improve the temperature coefficient of H_{ci}. This is very significant in view of the higher price of cobalt in recent times. The effort of various organic additives on the hot workability of melt-spun Nd-Fe-B magnets was reported by Iwasaki et al. (Ref 16). Magnequench has achieved an energy product as high as 52 MGOe (B_r = 14.5 kG) in MQ3 magnets on development basis.

5. Anisotropic Powder

The marketplace is demanding a higher energy product bonded magnet than those reported in Table 1. With the cross-licensing agreement between Magnequench, Mitsubishi Materials Corporation (MMTL), and Sumitomo Special Metals Corporation, hydrogen disproportionation desorption recombination (HDDR) anisotropic powder is becoming a commercial reality. Under this agreement, Magnequench will be the sole supplier of this powder (MQA-T). This will be the mainstream anisotropic powder from Magnequench. This process is

Table 4 Magnetic properties of anisotropic bonded magnets made from $Nd_{12.6}Fe_{Bal}Co_{11.6}B_6M_x$ HDDR powders

Addition M, at.%	Residual induction (B_r), kG	(bH_c), kOe	Intrinsic coercive force (H_{ci}), kOe	Maximum energy product ((BH)$_{max}$), MGOe
Ga 1.0	8.7	7.1	13.0	16.2
Zr 0.1	9.1	6.4	8.3	18.0
Nb 0.3	8.9	5.8	7.0	16.6
Hf 0.1	9.1	6.3	8.4	16.9
Ta 0.3	8.7	5.8	7.2	16.0

Table 5 MQA-T anisotropic powder

Magnetic characteristics (bonded magnet data)	
Residual induction (B_r), kG	8.2-8.6
Coercive force (H_c), kOe	7.0-7.5
Intrinsic coercive force (H_{ci}), kOe	12.5-14.5
Maximum energy product $(BH)_{max}$, MGOe	14.0-17.0
Temperature coefficient	
B_r to 100 °C, %/°C	−0.09
H_{ci} to 100 °C, %/°C	−0.55
Recommended magnetizing force (H_s), kOe	30
Physical properties	
Curie temperature (T_c), °C	420
Density, g/cm^3	6.1
Average particle size, µm	
Magnet manufacture	
Molding	Compression
Alignment field, kOe	24
Alignment orientation	Perpendicular
Die pressure, ton/cm^2	6

Note: Magnetic and physical properties are typical at room temperature.

shown schematically in Fig. 7. It utilizes the hydrogen-induced phase transformation as given below (Ref 17):

- $Nd_{16}Fe_{76}B_8$ (coarse-grained microstructure). Consists of basically three components: $Nd_2Fe_{14}B$, Nd-rich, $NdFe_4B_4$. This material is then exposed to hydrogen at 1 bar.
- $Nd_{16}Fe_{76}B_8H_{28}$. Three components: $Nd_2Fe_{14}BH_{2.7}$, Nd-rich hydride, $NdFe_4B_4$. Heat to 750 to 850 °C in hydrogen for 2 h.
- Fe, $NdH_{2.9}$, Fe_2B, $NdFe_4B_4$. Hold at temperature under vacuum for 1 h and cool to room temperature.
- $Nd_{16}Fe_{76}B_8$ (fine-grained microstructure). Consists of basically three components: $Nd_2Fe_{14}B$, Nd-rich, $NdFe_4B_4$.

The remanence of bonded magnets made using HDDR powders is dependent on the optimum addition of additives as given in Table 4 (Ref 18). The average grain size is ~0.3 µm and is nearly spherical in shape. Also there seems to be no second phase inside the crystalline grains, and the neodymium-rich phase is only observed in some parts of the grain boundary regions (Ref 18). The typical characteristics of bonded magnets are given in Table 5 and Fig. 8. It has fairly uniform magnetic properties over a wide range of particle sizes.

Magnequench introduced high-temperature bonded magnets to meet the market demand. They contain niobium as an additive. The bonded anisotropic magnet made using HDDR powder has an energy product of 15 MGOe, considerably

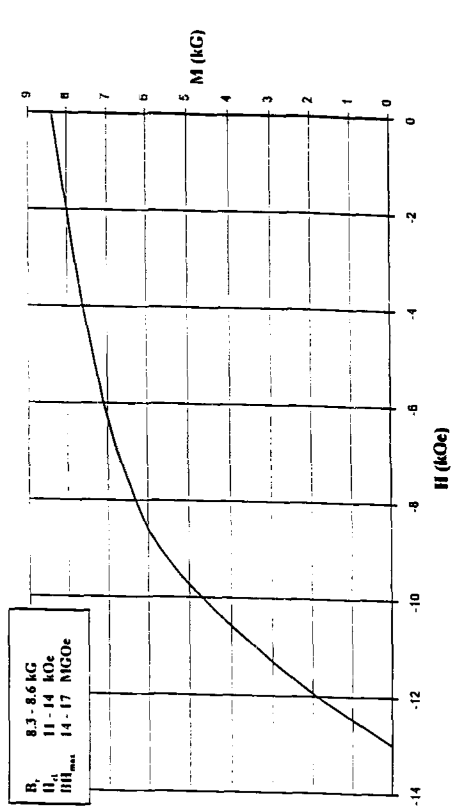

Fig. 8 Typical demagnetization curve of bonded magnet using HDDR powder

higher than those of bonded isotropic magnets. Also, high energy product MQ3 magnets were made possible by processing and alloy modifications.

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