High performance isotropic permanent magnet based on Nd-Fe-B

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We have fabricated a macroscopically isotropic permanent magnet material (Ovonic Hi-Rem™) which significantly exceeds conventionally understood performance limits for a material with saturation magnetization $M_s = 16$ kG. Enhanced magnetic properties, including a remanent magnetization $M_r = 9$–10 kG or more, and a maximum energy product in excess of 20 MGOe, are observed without preferred orientation. This material has been fabricated by rapid solidification of an alloy structurally and compositionally similar to $Nd_2Fe_{14}B$, but which depends on critical alloying additions and process parameters to obtain optimum magnetic performance.

The basis for the current understanding of conventional permanent magnets is the model of Stoner and Wohlfarth,[1] in which the material consists of an assembly of independent, noninteracting magnetic particles, each of which is uniformly magnetized and exhibits a single easy axis of magnetization (uniaxial). Assume that such a material is magnetically saturated by application of a very large magnetic field. Upon removal of the field, the magnetic moment of each individual particle will relax back (by rotation) to its particular easy axis. This yields a macroscopic value of the remanent magnetization M_r that is somewhat less than the saturation magnetization M_s if the particle easy axes are not perfectly oriented parallel to the applied field. In general,

$$M_r = M_s\langle|\cos(\theta)|\rangle, \tag{1}$$

where θ is the angle between the easy axis and the applied field, and $\langle\ \rangle$ indicates an ensemble average. For the special case where the particles are oriented at random, this yields $M_r = M_s/2$ for any direction of applied field. Special cases of Eq. (1) for a variety of different distributions of orientations have been derived,[2] and its direct verification for a real permanent magnet material has also been discussed.[3]

Those deviations from Eq. (1) that are conventionally seen in permanent magnets are for M_r smaller than predicted, due in part to inclusions of low-coercivity magnetic material. Larger values of M_r/M_s are possible in materials that exhibit high crystallographic symmetry (e.g., cubic), but these materials also exhibit very low values of coercivity H_c, making them unsuitable as permanent magnets. We are not aware of any previous examples of permanent magnet materials for which M_r/M_s significantly exceeds the value predicted from Eq. (1).

To assist in comparing our novel enhanced remanence magnet material (Hi-Rem™) to conventional permanent magnetic materials, we define a three-dimensional magnetic retention parameter Q defined by

$$Q = \sum_{x,y,z}\left(\frac{M_r}{M_s}\right)^2, \tag{2}$$

where this sum is taken over three orthogonal directions, in each case measuring M_r following saturation in the measurement direction. For an arbitrary distribution of orientations of noninteracting uniaxial particles [i.e., those obeying Eq. (1)], the value of Q can never be greater than 1. This follows rigorously from the observation that the values of M_r/M_s in the x, y, and z directions are, from Eq. (1), equivalent to the ensemble averages of the standard direction cosines a, b, and c such that $a^2 + b^2 + c^2 = 1$. Then

$$Q = \langle a\rangle^2 + \langle b\rangle^2 + \langle c\rangle^2 \leqslant \langle a^2 + b^2 + c^2\rangle = 1$$

as a consequence of the fact that the variance of a distribution (e.g., $\langle a^2\rangle - \langle a\rangle^2$) is always non-negative.

Two distributions of particular interest are the completely oriented case where $Q = 1$, and the completely non-oriented case where $Q = 0.75$. For the novel Hi-Rem™ material presented below, however, Q is substantially greater than 1 (see Table I), indicating a clear-cut violation of Eq. (1) and consequently of the assumptions of the Stoner–Wohlfarth model.

Recent developments[4] have focused on materials based on the tetragonal phase $Nd_2Fe_{14}B$,[5] which exhibits density 7.6 g/cm^3, Curie temperature $T_C = 590$ K, and room-temperature $M_s = 16$ kG (on pure single-crystal samples[6]). Magnets of this type have provided the highest magnetic performance to date,[7] and a phase of this type is the major component in the new magnetic material we have developed.[8,9]

In addition to conventional powder-metallurgy-based production techniques,[10,11] an alternative method of producing permanent magnets based on the $Nd_2Fe_{14}B$ phase

TABLE I. Measured values of magnetic parameters compared to literature values for related materials.

Sample	Easy axis $M_r/H_c/(BH)_{max}$ (kG/kOe/MGOe)	Hard axis $M_r/H_c/(BH)_{max}$ (kG/kOe/MGOe)	$Q = \sum_{x,y,z}(M_r/16\text{kG})^2$
376AV08(8)	10.7/8.0/18.5	9.5/7.6/15.5	1.2
469AA14(3)	10.7/>20/24.9	10.1/>20/22.1	1.3
Ref. 11	12.2/15/1/34.8	1/3/0.2	0.6
Ref. 13	8.2/14/14	isotropic	0.8
Ref. 15	7/14/13	isotropic?	0.6
Ref. 16	10.7/12.4/27	4.7/15.4/4.3	0.6

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makes use of rapid solidification.[12–15] In this process, the molten alloy is rapidly quenched on a rotating metal wheel (melt spinning) to produce a thin ribbon with isotropic fine-grained microstructure that may exhibit moderate magnetic performance in all directions, either as quenched[13] or after subsequent thermal processing.[14,15] Higher magnetic performance has been exhibited as anisotropic material manufactured from quenched ribbon using a die upset process.[16] Using the value $M_s = 16$ kG, both powder-processed and rapidly solidified magnets appear to be consistent with the independent-particle model.[17]

We have prepared rapidly solidified ribbon samples of a new permanent magnet material using a melt-spinning process.[8,9] The composition was based on $(Nd,Pr)_2Fe_{14}B$, with small but crucial alloying additions. Details of fabrication and analysis, including the significant differences from prior melt-spun materials of this type, will be presented in future publications.[9,18] The brittle ribbon (about 1 mm wide by 30 μm thick) fragmented into flakes about 5 mm long, each weighing on the order of 1 mg. Detailed magnetic measurements on two such ribbon fragments, 376AV08(8) and 469AA14(3), are presented below.

The samples were measured magnetically on an LDJ Inc. Model 9500 Vibrating Sample Magnetometer (VSM). The VSM has a conventional electromagnet capable of generating a field up to 20 kOe. The magnetic moment was detected to a resolution of less than 10^{-3} emu by centering the sample at the saddle point of a balanced set of four detection coils. The VSM was calibrated using a 2.4-mm-diam soft Ni sphere (NBS Standard Reference Material No. 772). Prior to each measurement it was necessary to premagnetize each sample individually in the appropriate direction using a pulsed magnetic field of about 120 kOe with a pulse width of 1 ms.

Each ribbon sample, after being weighed on a calibrated Cahn-21 microbalance with a 1-μg precision, was measured in each of three directions: the x direction parallel to the spin direction and to the long axis of the sample, the y direction perpendicular to the spin direction but still lying in the plane of the ribbon, and the z direction perpendicular to the plane of the ribbon. The premagnetized sample was saddled manually in zero applied field. The hysteresis loop was measured with a maximum field of 20 kOe, which was insufficient to fully reverse the initial magnetization. The magnetization is computed as $M = B - H = 4\pi\mu\rho/m$, where μ is the magnetic moment in emu, m the mass in grams, and a density $\rho = 7.4$ g/cm^3 was assumed. This is believed to be a lower estimate of the actual ribbon density. The value of the product $-BH$ was then computed around the loop, and the largest value taken as the maximum energy product $(BH)_{max}$.

The second quadrant demagnetization curves (M vs H) measured in the x, y, and z directions are shown in Fig. 1. for sample 469AA14(3). The z-direction demagnetization curve has been corrected (see Fig. 1) for the ribbon shape anisotropy, i.e., "sheared" to obtain M as a function of the internal field $H_{int} = H - NM$, where $N = 0.97$ is the demagnetization coefficient. This value for N is close to that predicted theoretically for an ellipsoid with dimensions similar to the flake dimensions.

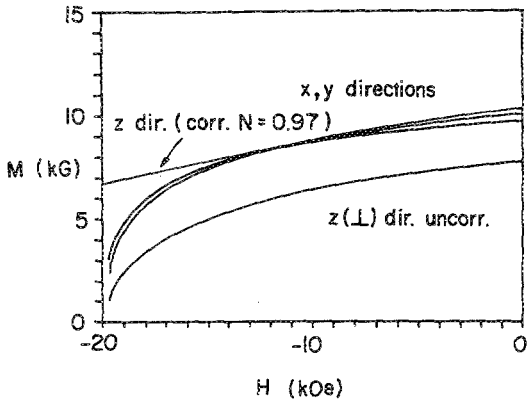

FIG. 1. Second quadrant demagnetization curve for sample 469AA14(3) measured in x, y, and z directions, and corrected z curve (using $N = 0.97$).

It is evident in Fig. 1. that after taking account of this shape anisotropy of the ribbon, the sample material itself is substantially isotropic, both in plane and perpendicular to the plane. This is consistent with the random orientation of the microscopic crystallites, which has been directly observed by both x-ray and electron diffraction.[8,9,18]

Table I presents a summary of magnetic data on two high-remanence ribbon samples (the coercivity H_c for sample 469AA14(3) was too large to be measured in our instrument), together with literature values for both isotropic and anisotropic magnetic materials. The "easy axis" here represents the direction exhibiting the largest value of remanence; the "hard axis" the smallest. The anisotropic materials[11,16] are assumed to have two equivalent hard axes, and the isotropic materials[13,15] three equivalent axes, in our estimates of the magnetic retention parameter Q. A value of $M_s = 16$ kG was assumed throughout. This is the standard value for $Nd_2Fe_{14}B$, and is consistent with preliminary measurements on several of our samples at high magnetic fields (55 kOe).

As the results in Table I indicate, the value of the magnetic retention parameter Q for the samples presented here are 1.2 and 1.3, both substantially greater than unity, in striking contrast with the conventional isotropic and anisotropic materials. We therefore conclude that our samples do not obey the Stoner–Wohlfarth model of noninteracting particles.

More detailed x-ray analyses and microstructural studies have been carried out,[8] and their relation to the enhanced magnetic performance will be discussed in future publications.[9,18] In addition, we are developing a phenomenological model that may account for this enhancement, and this too will be presented in the future.[18]

From the point of view of potential applications, the enhanced isotropic magnetic performance observed in single flakes can be maintained in compacted magnetic bodies, and the compaction process is simplified by the isotropic nature of the material, which remains isotropic after compaction.

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