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DETERMINATION OF THE DEGREE OF CRYSTALLITES ORIENTATION IN PERMANENT MAGNETS BY X-RAY SCATTERING AND MAGNETIC MEASUREMENTS

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Abstract - Permanent magnet compounds are basically ferromagnets with uniaxial anisotropy. In different cases, the magnets are elaborated by sintering of powders previously oriented and compressed under field. The value of the remanent induction \( B_r \) is directly dependent on the degree of crystallites alignment, and we present in this paper two different quantitative methods to determine it. X-ray analysis involves the comparison of the intensities of Bragg reflexions, for different values of the angle of a direction \([hkl]\) with respect to the unique crystallographic axis of the compound. Magnetic analysis involves the comparison between experimental magnetization curves and calculated ones for different statistical distributions of crystallites orientations. Both methods were applied to Nd-Fe-B and SmCo\(_5\) magnets. They reveal that the misorientation of particles in these systems leads to a reduction of the remanent induction by about 10%.

INTRODUCTION

Permanent magnet compounds are basically ferromagnets with uniaxial anisotropy. The coercivity of the magnets, closely related to this anisotropy, develops in materials which are inhomogeneous at a microscopic scale. Such a property is in particular obtained in ferrites and rare earth-transition metal magnets, by sintering of powders previously oriented and compressed under field. The value of the remanent induction \( B_r \) is directly dependent on the degree of crystallites alignment. A quick method to determine an average value of their degree of alignment consists in measuring the remanent induction \( B_r \) of the magnet with respect to the spontaneous induction of the compound \( /I/ \). We present in this paper a more quantitative analysis of the statistical distribution of grains orientation, which was obtained by X-ray analysis and magnetostatic measurements, on NdFeB and SmCo5 magnets.

X-RAY DETERMINATION OF THE DEGREE OF GRAIN ALIGNMENT

For a reflection \( [hkl] \), Bragg scattering occurs if the direction \([hkl]\) is along the scattering vector \( \vec{K} \), defined as the bissectrice of the angle between the direction of the incident beam and that of the scattered beam. In a polycrystalline sample, in

*partly supported by a fellowship from "Aimants UGIMAG S.A.", France
Fig. 1 - X-ray diffractogram of a Nd-Fe-B magnet obtained with the Co radiation ($\lambda_{Co} = 1.7903$ Å).

which crystallites orientations are perfectly at random, the probability for any direction [hk1] to be along $K$ is constant. In the case of a single crystal, it is necessary to adjust the crystal orientation in order to align [hk1] along $K$. Permanent magnets elaborated by sintering of powders, can be considered as an assembly of single crystals, for which the crystallographic $c$-axes, i.e. the directions [001] of the reciprocal space, are distributed within a certain solid angle. If the axis of the solid angle, $z$, is aligned along $K$, the probability $p(\omega//)$ for crystallographic $c$-axes to be along a given direction at an angle $\omega//\text{with respect to } z$ is simply equal to the probability for a direction [hk1] at an angle $\omega_{hk1} = \omega//$, from [001] to be along $K$. Thus the comparison of the intensity of a reflection [hk1], $I_{hk1}$, magnet, to that observed in a polycrystalline sample, $I_{polY}$, is a measurement of the proportion of crystallites along any direction at $\omega//\text{from } z$.

We used this method to determine the degree of grain alignment in Nd-Fe-B and SmCo$_5$ magnets. X-ray diffractograms were recorded on slices of Nd-Fe-B permanent magnets, cut at different thicknesses perpendicularly to $z$. The results obtained show that the degree of grain alignment is a constant over the volume of the magnet, except on a very thin skin of about 0.1 mm at the surface. Typical X-ray pattern is presented in figure 1. Comparison to the intensities measured on a non-oriented powder /2, 3/ leads to the determination of $I_{hk1}$ in figure 1. As shown in figure 3, the decrease of $I_{hk1}$ in figure 2.

Fig. 2 - $I_{hk1}/I_{polY}$ as function of $\omega_{hk1}$ between directions [hk1] and [001].

*the sample of SmCo$_5$ was kindly provided to us by "Aimants UGIMAG S.A."
with $\omega$ is approximately a

\[
\text{gaussian } \exp(-\omega^2/2\sigma^2) \text{ with } \sigma = 18.7^\circ.
\]

This would mean that

99% of crystallites make an

angle smaller than 55° with

respect to the $z$-axis of the

magnet. However, the

observation of weak

reflections with $\omega// > 45^\circ$

reveals an additional small

contribution of another

gaussian with $\sigma' = 36^\circ$,

corresponding to a much

broader distribution of

crystallites orientation

(Figure 3). The proportion of

such crystallites is

approximately 15% of the

total number of crystallites.

This property can be

understood by considering

that a proportion of

crystallites obtained after

grinding of the bulk ingots

are not single crystals. Such

crystallites are not efficiently oriented in the

field applied during the

compression.

The same analysis was performed for a SmCo$_5$ magnet. The results are presented in

figures 2 and 3. Very similar results are obtained. The main gaussian describing the

statistical distribution of grains orientation is determined by $\sigma = 14^\circ$. The

accuracy of the results is not sufficient to reveal a possible additional contribu-

tion from another gaussian describing a broader distribution of grains orientation.

Such a contribution is in any case much weaker than in NdFeB, and suggests therefore

that, in SmCo$_5$ magnets, almost all grains can be considered as single crystals.

III - MAGNETIC DETERMINATION OF THE DEGREE OF GRAINS ALIGNMENT

The field dependence of the magnetization of an uniaxial single-crystalline compound

has been calculated by Néel et al./4/ for any angle $\theta$ between the applied field and

the easy $c$-axis. The first magnetization curve can be divided into two parts

associated with different physical processes. In low fields, the crystal is

decomposed into several domains, and the magnetization measured along the field $H$

is

\[
m(\phi, H) = M_s \left[ \cos^2 \phi / H_D + \sin^2 \phi / (H_D + H_A) \right]
\]

(1)

where $M_s$ is the spontaneous magnetization, $H_D$ the demagnetizing field for saturated

magnetization and $H_A$ the anisotropy field. The magnetization follows this law until

a threshold field, $H_g(\phi)$, for which the crystal becomes single domain. Above $H_g$, the

angle $\theta$ between the magnetization and the easy axis is given by:

\[
(H_A/2H) \sin 2\phi + \sin (\theta - \phi) = 0
\]

(2)

and the magnetization measured along the field is:

\[
m(\phi, H) = M_s \cos (\phi - 0)
\]

(3)

The calculated recoil magnetization curve, assuming that the coercivity impedes the

nucleation of reverse domains, corresponds to the second process alone.

As described in the previous section, the permanent magnets considered here are

formed of independent crystallites whose easy axes are distributed about a preferred

direction $z$. When the field is applied along $z$, crystallites which make the same

angle $\omega// with respect to $z$, i.e. with respect to the field, exhibit identical

magnetization law. The number of crystallites between $\omega// and $\omega// + \text{d}\omega// is obtained

by integrating over $\alpha$ around the $\frac{2}{2}$-axis:

\[
\text{...}
\]
where $p(\omega_//)$ is the statistical distribution of crystallites. When the field is applied perpendicular to $z$, the crystallites behaving identically make the same angle $\omega$ with respect to the field. The number of crystallites between $\omega_\perp$ and $\omega_\perp + d\omega_\perp$ is:

$$N_\perp(\omega_\perp) d\omega_\perp = \int_{\alpha=0}^{2\pi} \sin \omega_\perp p(\omega_//) d\omega_// \ d\alpha$$

with $\omega_\perp$ depending on $\alpha$ through:

$$\cos \omega_\perp = \sin \omega_// x \cos \alpha$$

In both cases, the calculated magnetization of the magnet is therefore:

$$M(H) = \frac{\int_{0}^{\pi/2} N_\perp(\omega_\perp) x m(\omega_\perp, H) d\omega_\perp}{\int_{0}^{\pi/2} N_\perp(\omega_\perp) d\omega_\perp}$$

where $m(\omega, H)$ is deduced from relations (1) to (3) with $\omega_\perp = \omega_// \text{ or } \omega_\perp$ depending whether the field is along or perpendicular to $H$.

Subsequently, this method was used to deduce the degree of grains orientation from magnetic measurements. The field dependence of the magnetization for a Nd-Fe-B magnet, under fields applied respectively along $z$ and perpendicular to $z$, are presented in figure 4. Satisfactory agreement between calculated first magnetization curves and experimental results can be obtained if considering a unique gaussian distribution for $p(\omega_//)$. However a better fit is obtained with the superposition of two gaussians with $\sigma = 17^\circ$ and $\sigma' = 28^\circ$, and respective weight 90 % and 10 % (Figure 4).

The differences between experimental and calculated magnetization curves in intermediate fields can be attributed to the inhomogeneities of the demagnetizing field in the cubic specimen studied, as verified by measuring an Fe sample of identical shape.

A same analysis for SmCo$_5$ magnet is presented in figure 5. The calculated curves were obtained for a main (95 %) gaussian with $\sigma = 15^\circ$, and a weak additional contribution (5 %) with $\sigma = 30^\circ$. 

![Fig. 4 - Experimental (---) and calculated (—) magnetization curves of a NdFeB magnet. Calculation parameters : $M_0 = 150$ e.m.u./g, $H_A = 73$ kOe.](image1)

![Fig. 5 - Experimental (---) and calculated (—) magnetization curves of a SmCo$_5$ magnet. Calculation parameters : $M_0 = 98$ e.m.u./g, $H_A = 350$ kOe.](image2)
IV - CONCLUSION

Crystallites orientation in permanent magnets was determined by two different methods involving X-ray and magnetic analysis respectively. Consistent results were obtained. The reduction of the remanent induction $B_r$, due to misalignment of the grains, reaches about 10% in both Nd-Fe-B and SmCo$_5$ magnets. Comparing both methods, X-ray analysis is more quantitative, but not as easy to realize as magnetic analysis. It is worth recalling that the usual way to determine grains orientation involves a direct measurement of $B_r$. The result may be affected by a possible decrease of the induction resulting from domain nucleation in grains of small coercivity. The measurement of the first magnetization curve considered in this study, is obviously not affected by such a phenomenon. Finally, a straight generalization of this method would permit a more quantitative analysis of magnetic properties in uniaxial systems where single crystals are not available /3,5/.

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