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ScienceDirect
Journal of Magnetism and Magnetic Materials 320 (2008) e40–e42
Journal of magnetism and magnetic materials
www.elsevier.com/locate/jmmm

Determination of the crystallographic texture of sintered PrFeB magnets based on X-ray diffraction patterns

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Available online 19 February 2008

Abstract

An alternative method for determining the degree of crystallographic alignment ($\langle \cos\Theta \rangle$) of the magnetic $Pr_2Fe_{14}B$ phase (Φ) is proposed. The method is based on the relative X-ray diffracted intensities of the Φ planes of sintered magnets with and without texture. The degree of crystallographic alignment is also determined by X-ray pole figures using the (0 0 4) reflection and considered as a standard reference for comparison. The method is applied to $Pr_{16}Fe_{76}B_8$ magnets with $0.51 \leqslant \langle \cos\Theta \rangle \leqslant 0.97$. The difference between the crystallographic alignments determined by these two methods is 3% within the experimental error. The advantages and limitations of using X-ray diffraction patterns to quantitatively evaluate the texture of sintered magnets are also discussed.

PACS: 71.20.Eh; 61.10.Nz

Keywords: Crystallographic texture; Hydrogen decrepitation; PrFeB; X-ray diffraction; X-ray pole figure

1. Introduction

X-ray diffraction (XRD) has been applied successfully to estimate the degree of crystallographic texture in permanent magnets. The quantitative analysis of NdFeB and SmCo magnets was first discussed by Givord et al. [1]. The statistical distribution of grain alignment was evaluated by plotting the ratio of the intensity of a reflection ($h\,k\,l$) of an anisotropic magnet to that observed in an isotropic sample versus ψ (angle between a certain ($h\,k\,l$) plane and a (0 0 l) plane). Zhou et al. [2] also estimated the alignment of sintered NdFeB magnets by correlating the intensity of the diffracted (0 0 6) peak with remanence. Ferrante et al. [3] evaluated the magnetic alignment of hot-rolled (Nd,Pr)FeB magnets, also using this technique. More recently, it has been shown that the alignment index $\langle \cos\Theta \rangle$, determined by the (0 0 4) X-ray pole figures (XRPF) analysis, is ideal to represent the degree of alignment of PrFeB magnets [4–6]. This index is convenient for use in mathematical equations for quantitative calculations of remanence values [5]. In the present study, a method for obtaining $\langle \cos\Theta \rangle$ is proposed, based on data from the relative diffracted intensities of the Φ phase planes on the X-ray diffraction pattern (XRDP) of anisotropic and isotropic magnets. A comparison is also made of the $\langle \cos\Theta \rangle$ obtained using XRDP and XRPF.

2. Experimental

Sintered $Pr_{16}Fe_{76}B_8$ magnets were prepared using the powder metallurgy route together with the hydrogen decrepitation (HD) process [5]. The degree of alignment ($0.51 \leqslant \langle \cos\Theta \rangle \leqslant 0.97$ [5]) was varied by changing the milling time. The isotropic magnet was obtained without applying the orienting magnetic field. The crystallographic alignment was initially determined by XRPF, using the (0 0 4) reflection [5,6] considered a standard reference for a comparison with XRDP. XRD patterns were obtained with Cu Kα radiation on an orthogonal magnet surface in the direction of the orienting field. The 2θ angle shifted from 20° to 70° at a scanning rate of $1^\circ\,min^{-1}$.

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doi:10.1016/j.jmmm.2008.02.012

3. Results and discussion

3.1. The method

Table 1 lists the ratio of the normalized intensities of the matrix phase in magnets with texture (I) to the normalized intensities of the isotropic Φ phase (I_0), which was determined from the XRDP. This procedure was carried out for planes whose angle versus a (0 0 l) plane (ψ) varied from 0° to 90° (0–1.57 rad). Linear, exponential, Lorentzian and Gaussian functions were used to test the best fit for the experimental data, showing a correlation coefficient (R^2) of 0.92, 0.95, 0.96 and 0.97, respectively. Gaussian and Lorentzian regressions were close due to the similarity of these two mathematical expressions. The former was chosen for the orientation distribution of the (0 0 l) planes. Fig. 1 shows this best fit for the $Pr_{16}Fe_{76}B_8$ magnet presented in Table 1.

To determine $\langle \cos\Theta \rangle$ using the XRD patterns, the Gaussian function was integrated using the expression [7]:

$$\langle \cos\Theta \rangle = \frac{\int_0^{\pi/2} f(\alpha)\,\sin(\alpha)\,\cos(\alpha)\,d\alpha}{\int_0^{\pi/2} f(\alpha)\,\sin(\alpha)\,d\alpha} = \frac{A}{B}, \quad (1)$$

Table 1
Normalized intensities of planes for the isotropic (I_0) and anisotropic (I) $Pr_2Fe_{14}B$ phase

Plane	ψ (°)	I_0 (%)	I (%)	$(I/I_0)\times 100$
(0 0 4)	0	20	35	175
(1 0 5)	15	70	100	143
(2 0 4)	35	50	45	90
(2 2 4)	45	80	39	49
(2 2 2)	64	50	30	60
(3 1 1)	78	80	29	36
(4 1 0)	90	100	39	39

The anisotropic magnet presents $\langle \cos\Theta \rangle = 0.71 \pm 0.02$, determined by XRPF.

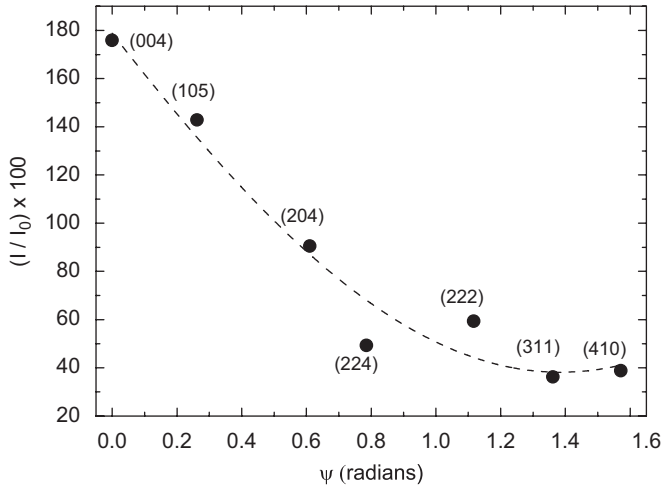

Fig. 1. Ratio of the experimental normalized intensity (of an oriented sample) to the relative intensity of a sample with no texture as a function of ψ. The planes are identified.

where α is the angle between the tetragonal c-axis and the orienting field. Eq. (1) was solved using a commercial mathematical software program with two decimal places. Table 2 presents the values of the parameters A, B and $\langle \cos\Theta \rangle$ obtained by XRDP. Highly coherent values of $\langle \cos\Theta \rangle$ were found, as will be shown in the next section. However, it should be noted that the function established from the Gaussian distribution for the curve shown in Fig. 1 must not be integrated up to $\pi/2$ (90°). The minimum relative intensity was verified for $\psi \approx 1.38$ rad (78°). This value, in radians, must be used as the upper integration limit in Eq. (1). From this point onwards, the data are physically meaningless since the intensity increases probably due to diffraction of crystalline planes that show a small angular deviation from those that satisfy the Bragg condition. If the integration is carried out up to $\pi/2$, a smaller crystallographic texture of the $Pr_2Fe_{14}B$ phase will be obtained than that obtained with XRPF. The integration of Eq. (1) with $\pi/2$ as the upper integration limit should be used only if the intensity does not reach a minimum before $\pi/2$. Therefore, the upper integration limit is variable and depends on the degree of crystallographic alignment (the same approach is valid for XRPF). This is clearly illustrated by the curves of magnets with different degrees of alignment depicted in Fig. 2.

3.2. Comparison of XRDP and XRPF

Fig. 3 compares the crystallographic alignments obtained by XRDP and XRPF. As can be seen, the values showed an excellent agreement, with a variation of about 3%. In each case, the correlation coefficient for the XRDP regressions (R^2_{XRDP}) is higher than 0.95, indicating the reliability of the proposed method.

3.3. Why does XRDP substitute XRPF?

The pole figure method uses the reflection of a parallel plane to the surface of the sample and orthogonal to the easy magnetic axis in order to characterize the crystallographic alignment. A (0 0 l) plane is normally used. The position of the sample is tilted at two different angles ($0° \leqslant \alpha \leqslant 90°$ and $0° \leqslant \beta \leqslant 360°$) to evaluate the degree of orientation distribution of the cited plane. Considering that the distribution of any crystalline plane will present a radial symmetry due to the aligning magnetic field, XRD and XRPF analyses act similarly. There is no need to use β

Table 2
Calculation parameters and alignment index $\langle \cos\Theta \rangle$ of $Pr_{16}Fe_{76}B_8$ magnets

Sample	A	B	$\langle \cos\Theta \rangle$
#1	37.35	73.63	0.51
#2	37.18	53.29	0.70
#3	25.14	30.31	0.83
#4	8.72	9.11	0.96

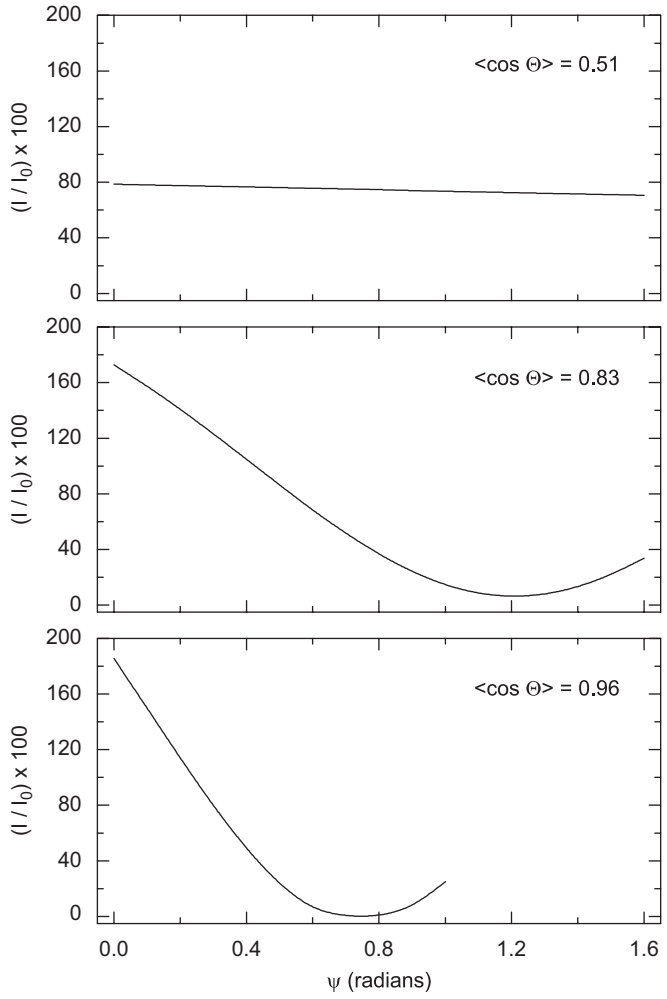

Fig. 2. Ratio of the experimental normalized intensity (for magnets with several degrees of alignment) to the relative intensity of a sample with no texture as a function of ψ.

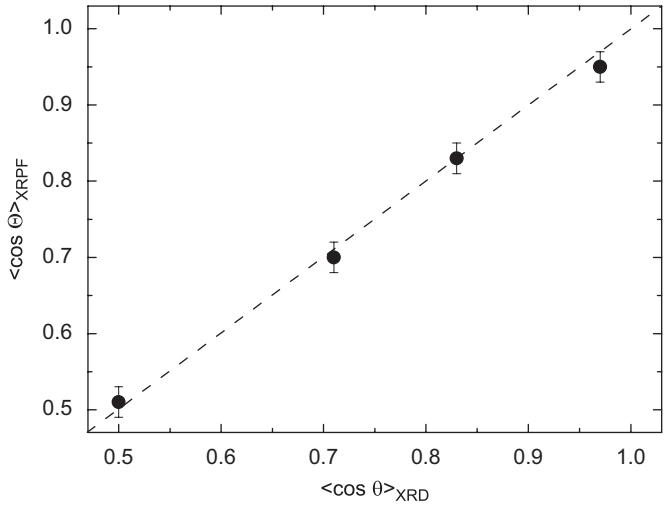

Fig. 3. Comparison of the crystallographic alignments obtained by XRPF and XRDP.

due to symmetry and α is substituted by the interplanar angle ψ.

3.4. Advantages and limitations

XRDP, compared to the XRPF technique, presents the following features:

- The crystallographic alignment of any magnetic phase in a magnet can be obtained. The crystalline planes of the phase must be correctly identified and must necessarily show a distribution with a radial symmetry;
- The XRDP analysis can be four times faster than XRPF, essential when monitoring $\langle \cos\Theta \rangle$ during magnet production;
- No special arrangement is necessary to perform XRDP compared to XRPF; and
- The determination of $\langle \cos\Theta \rangle$ by XRD patterns is easier than XRPF because there is less data to process.

4. Conclusions

An alternative method for determining the degree of crystallographic alignment of the magnetic $Pr_2Fe_{14}B$ phase in sintered magnets was proposed. For sintered magnets with $0.51 \leqslant \langle \cos\Theta \rangle \leqslant 0.97$, the difference between the crystallographic alignments determined by XRD patterns and XRPF analyses was 3% within the experimental error. The crystallographic alignment of any magnetic phase in a magnet can be obtained. However, the crystalline planes of the phase must be distributed with a radial symmetry.

Acknowledgments

The authors thank IPEN-CNEN and FAPESP for their funding of this investigation and for providing a research grant (E.A. Périgo, Contract no. 2005/04711-2). The authors are also indebted to Rene R. de Oliveira for the XRD analyses.

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