Materials Science Forum Vols. 660-661 (2010) pp 139-144
Online available since 2010/Oct/25 at www.scientific.net

doi:10.4028/www.scientific.net/MSF.660-661.139

DETERMINATION OF THE CRYSTAL ALIGNMENT OF Pr-Fe-B MAGNETS USING THE (105) X-RAY POLE FIGURE

Castro, A.R.M[1,a], Takiishi, H[1,b], Faria, R.N[1,c], Lima, N.B[1,d]
[1]Energy and Nuclear Research Institute, Av. Lineu Prestes 2242 - CEP: 05508-000
São Paulo - Brazil
[a]arcastro@ipen.br, [b]takiishi@ipen.br, [c]rfaria@ipen.br, [d]nblima@ipen.br

Keywords: Magnets; Texture; PrFeB-alloys

Abstract. The crystallographic alignment of various permanent magnets has been investigated by X-ray pole figure analysis. Attempts have been made to measure the degree of alignment of these sintered magnets using the (105) reflection. It has been shown that the (105) pole figure can be used only to verify small differences in texture in magnets high degree of crystallographic alignment. A comparison between the measured and the calculated L_{105} index showed good agreement.

Introduction

It is well known that there is a direct correlation between remanence and energy product with texture or the crystal alignment of permanent magnets. Therefore, several studies to evaluate the degree of crystal alignment of magnets, using X-ray diffraction, have been reported [1-9]. Recently, a systematic study of magnetic alignment in Pr-Fe-B sintered magnets using the (004) reflection have been reported [10]. These Pr-based magnets were produced using the hydrogen decrepitation process [11]. It has also been reported that the (105) reflection is not appropriated to sintered magnets with low degree of crystallographic alignment [10]. In this study, the crystal alignment of $Pr_{16}Fe_{76}B_8$ permanent magnets has been further investigated using the (105) X-ray pole figure analysis. The degree of alignment in sintered magnets was carried out by changing the milling time and the intensity of the orientation field [12, 13]. Hydrogen desorption from the powder prior to the milling step was also employed to vary the degree of alignment in the magnets [14].

Experimental

A commercial $Pr_{16}Fe_{76}B_8$ alloy in the as-cast state was used in this study. To produce the sintered Pr-based magnets using the hydrogen decrepitation (HD) process [11], 35 g of the bulk ingot were placed in a stainless steel hydrogenation vessel, which was then evacuated to backing-pump pressure. Hydrogen was introduced to a pressure of 1 bar, which resulted in decrepitation of the bulk material. The standard decrepitated hydride material was then transferred to a “roller” ball-mill under a protective atmosphere and milled for several hours (18, 27, 36 and 45 hours) using cyclohexane as the milling medium. The resultant fine powder was then dried for 1 h and transferred to a small cylindrical rubber tube under a nitrogen atmosphere. The fine powder was aligned by pulsing three times to a 6 T magnetic field, pressed isostatically at 1000 kg cm^{-2} and then vacuum sintered for 1h at 1060°C, followed by cooling on the furnace (~3.5°C min^{-1}). Annealing was carried out under vacuum at 1000°C for 24 hours. Magnetic measurements of the HD sintered magnets were performed in a permeameter after saturation in a pulsed field of 6 T.

Partially desorbed (PD) powder was prepared from the decrepitated hydride by a heat treatment in vacuum at 300°C for 5 hours. Total desorption (TD) of the powder was also carried out in vacuum but at 600°C for 5 hours. The partially and totally desorbed material was then processed in the same manner as the standard decrepitated hydride (HD) material. The samples, as-sintered and annealed (A), were examined by X-ray diffraction techniques.

Measurements of (105) and (004) pole figures were carried out by the Schulz's reflection method with a diffractometer, using the CrKα radiation [1]. The tilt angle (α) was varied from 0^o to 75^o in steps of 5^o. The angle of rotation, azimuth angle (β), was varied from 0^o to 360^o in steps of 5^o. The intensities of the pole figure data were normalized by:

$$I(\alpha_j) = \frac{\sum_{i=1}^{72} f(\alpha_j, \beta_i)}{72.I(\alpha_1)} \qquad j = 1,16 \qquad (1)$$

The (105) normalized intensity data was fitted for two Lorentz functions centered at 0^o and 15^o from magnet alignment direction and the ratio L_{105} was calculated by:

$$L_{105} = \frac{L(\Theta_c = 15^o)}{L(\Theta_c = 0^o)} \qquad (2)$$

$L(\Theta_c = 15^o)$ and $L(\Theta_c = 0^o)$ are the maximum intensity values of the Lorentz functions.

Results and Discussion

Normalized intensities for the reflections (105), obtained using equation (1) and as a function of α angle for three permanent magnets processed using different milling times, in the as-sintered, are shown in Figure 1. Sintered $Pr_{16}Fe_{76}B_8$ magnets prepared using longer milling time (samples HD 27 and 36) showed better a sharper curve pattern whereas the magnet produced using shorter milling time (HD18) exhibited a broader pattern and no index L_{105} could be determined in this case.

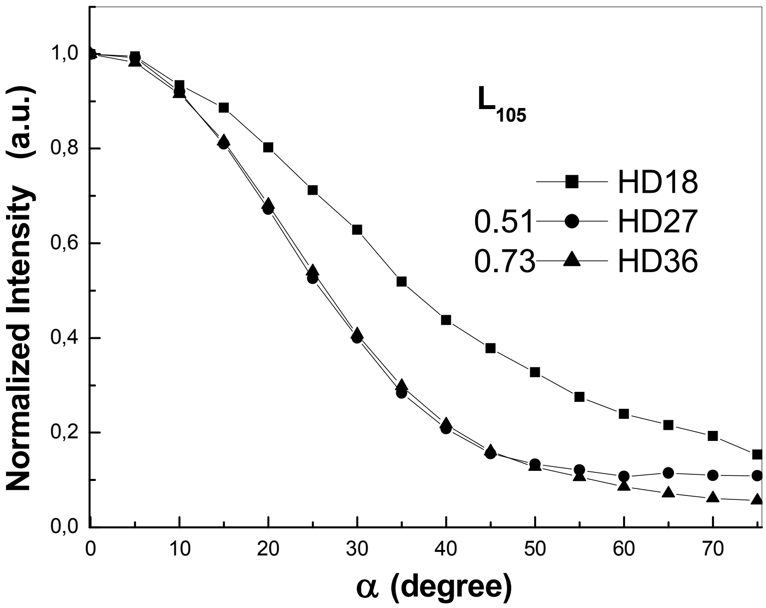

Fig. 1 - Normalized intensities as a function of angle α for various magnets in the as-sintered state, for the reflections (105).

Normalized intensities for the reflections (105) the previous permanent magnets in the annealed state, are shown in Figure 2. Again, sintered $Pr_{16}Fe_{76}B_8$ magnets prepared using longer milling time (samples HD 27A and 36A) showed better a sharper curve pattern than that of the magnet produced using shorter milling time (HD18A). Once again, the index L_{105} could not be determined for this case. Some improvement has been obtained with annealing at high temperature.

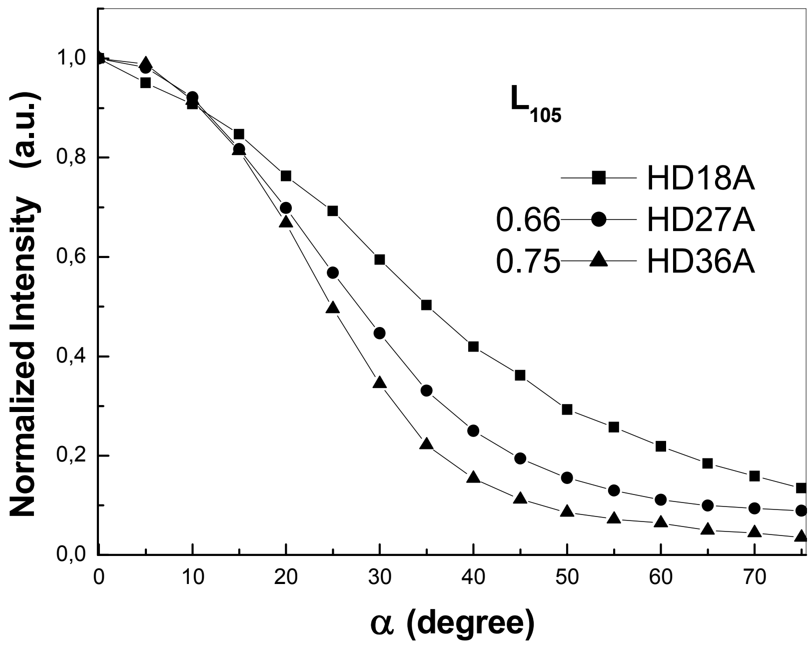

Fig. 2 - Normalized intensities as a function of angle α for various magnets in the annealed state, for the reflections (105).

Curves for normalized intensities (105) plotted as a function of α angle for $Pr_{16}Fe_{76}B_8$ magnets with high degree of alignment are shown in Figure 3. These three as-sintered magnets showed almost identical curves with sharp pattern peculiar to samples with high degree of alignment. Normalized intensities for these three magnets in the annealed state are shown in Figure 4. In this case, however, they exhibited a maximum at approximately 15^o for two magnets (HD45A and TD18A), due to the influence of the [001] magnetic alignment direction. This shows that these two magnets are slightly better aligned than that which did not show a maximum at low angles. Annealing also improved further the degree of crystallographic alignment of the as-sintered $Pr_{16}Fe_{76}B_8$ magnets that already had a high degree of alignment.

Figure 5 shows a correlation of remanence and the index L_{105}, data taken from Ref. [10] for the $\langle\cos\Theta\rangle$. The flat line or saturation for the $\langle\cos\Theta\rangle$ above 0.96 shows a low sensitivity in this region. Although the L_{105} index is very sensitive in the case of magnets with a high degree of alignment, it showed a wide scatter and no meaningful conclusion could be drawn when considering $Pr_{16}Fe_{76}B_8$ magnets in the as-sintered and annealed conditions. Thus, the index L_{105} proved not to be suitable to represent permanent magnets with low and medium degree of crystallographic alignment.

As reported previously [10], the index $\langle\cos\Theta\rangle$ determined by the (004) pole figure analysis is a good index to represent the degree of alignment of sintered $Pr_{16}Fe_{76}B_8$ magnets. However, the $\langle\cos\Theta\rangle$ obtained by pole figure analysis was found to be less sensitive at high degrees of alignment because of the I(α) broadening [10]. Thus, in sintered $Pr_{16}Fe_{76}B_8$ magnets with high degree of crystallographic alignment the index L_{105} can detect small differences in texture which is not possible with the index $\langle\cos\Theta\rangle$. This can be the case in a magnet industrial production line where all magnets are fabricated with a high degree of crystal alignment and a more accurate measurement is necessary. Calculated L_{105} index of crystallographic alignment for the various permanent $Pr_{16}Fe_{76}B_8$ magnets was also included for a comparison, as shown in Figure 6. Some disparity is only found for low values of L_{105} but, in general, good agreement was observed for the other samples.

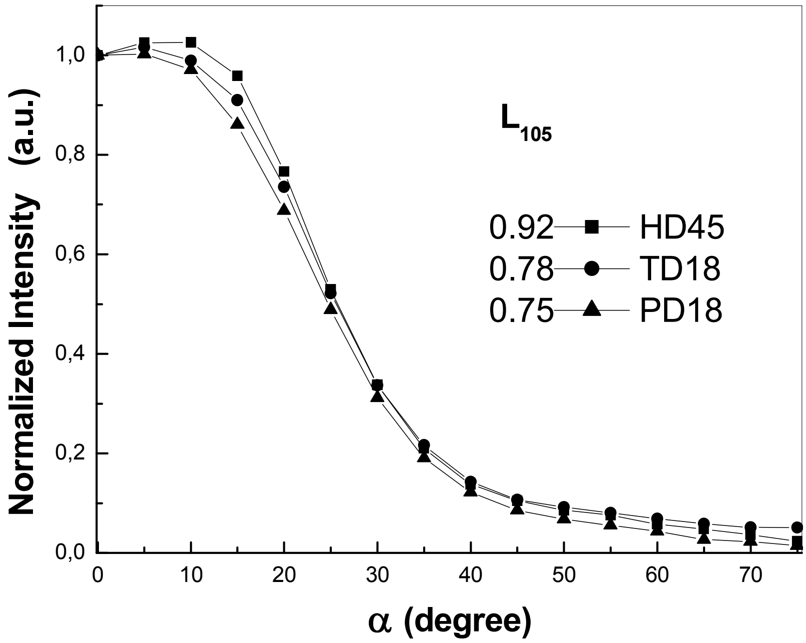

Fig. 3 - Normalized intensities as a function of angle α for three magnets in the as-sintered condition and with high degrees of alignment, for the reflections (105).

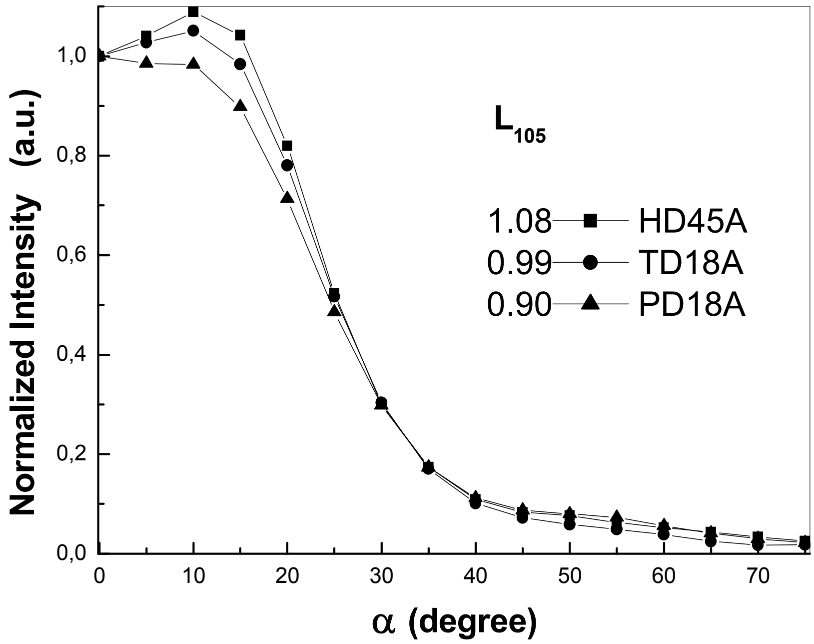

Fig. 4 - Normalized intensities as a function of angle α for three annealed magnets with high degrees of alignment, for the reflections (105).

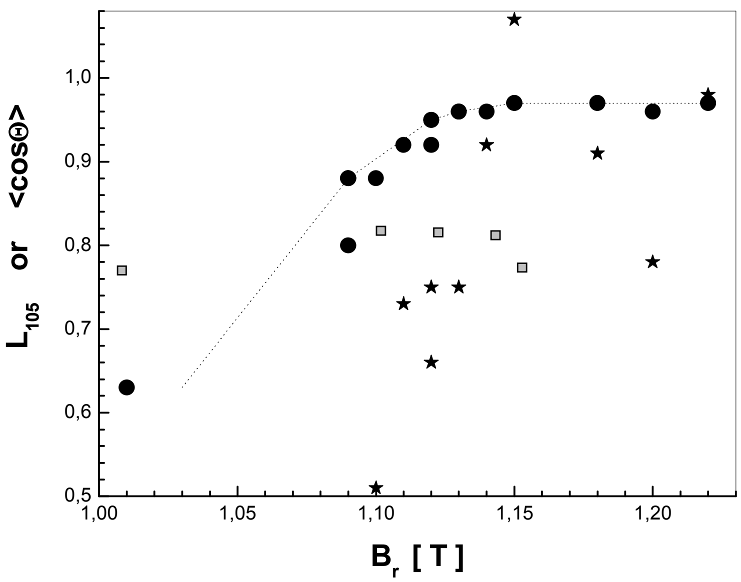

Fig. 5 Correlation of B_r with L_{105} (*) and <cosΘ> data (•) from Ref. [10].

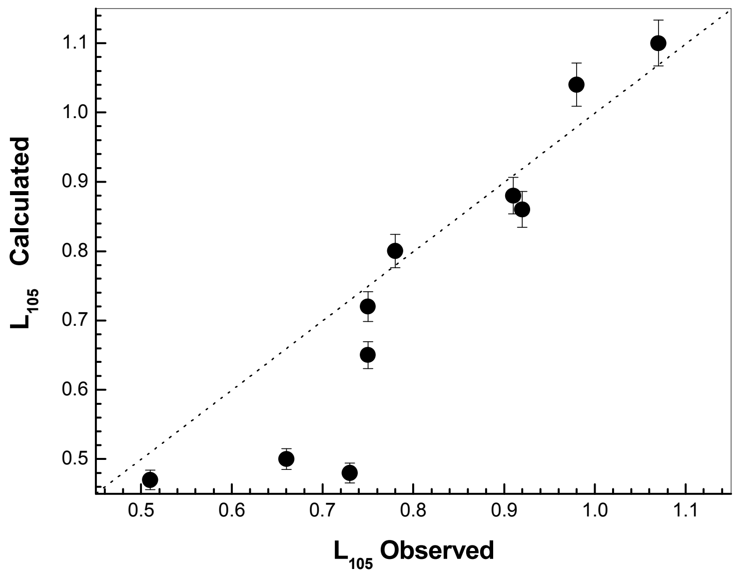

Fig. 6 Correlation of L_{105} observed with L_{105} calculated.

Magnetic domains of $Pr_{16}Fe_{76}B_8$ sintered magnets have been observed by Kerr effect and histogram of the angular distribution of domain alignment has been used to determine the magnetic texture [15]. It has been shown that the Kerr technique, although time-consuming, can provide a reasonable estimate for magnets with low and medium degrees of crystal alignment. Thus, this technique is inappropriate for magnets with high degree of crystallographic alignment. This behavior can be easily understood by realizing that it becomes very difficult to determine the crystallographic alignment using the Kerr technique in sintered permanent magnets with very fine grain sizes. The origin of (105) reflection of X-ray diffraction and quantitative evaluation of the alignment degree for sintered Nd-Fe-B magnets have also been reported previously [16].

Conclusions

In $Pr_{16}Fe_{76}B_8$ magnets with a high degree of crystallographic alignment, the index L_{105} could be employed to verify small differences in texture. A comparison of the index L_{105} with the index <cosΘ>, also determined by the (004) pole figure analysis; showed that, in general, the latter is a good index to represent the degree of alignment of $Pr_{16}Fe_{76}B_8$ sintered magnets but with low sensitivity for highly aligned samples. If industrial magnets with high degree of alignment are to be tested the L_{105} should be used instead of the index <cosΘ>. Kerr effect can be used for magnets with low degree of crystal alignment. Good agreement has been achieved between the observed and the calculated L_{105} index.

Acknowledgements
Many thanks are due The State of São Paulo Research Foundation and Energy and Nuclear Research Institute for supporting this investigation. Thanks are also due to M. M. Serna and E. Galego for their help during the course of this work.

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Advanced Powder Technology VII

doi:10.4028/www.scientific.net/MSF.660-661

Determination of the Crystal Alignment of Pr-Fe-B Magnets Using the (105) X-Ray Pole Figure

doi:10.4028/www.scientific.net/MSF.660-661.139