Materials Science Forum Vols. 408-412 (2002) pp. 173-178

Neutron Diffraction Texture Study of NdFeB As-Cast Alloys and Magnets

S. Rivoirard[1], D. Chateigner[2], P. de Rango[3] and D. Fruchart[3]

[1] Consortium de Recherche pour l'Emergence des Technologies Avancées, CNRS, BP 166, FR-38042 Grenoble, France

[2] Laboratoire de Cristallographie et Sciences des Matériaux, ISMRA, 6 Bd. du Maréchal Juin, FR-14050 Caen, France

[3] Laboratoire de Cristallographie - CNRS, BP 166, FR-38042 Grenoble, France

Keywords: As-Cast Texture, Extrinsic Magnetic Anisotropy, Hot Deformation Texture, NdFeB Magnets

Abstract. Macroscopic magnetic anisotropy in NdFeB permanent magnets results from the hot forging process of NdFeB bulk alloys. This process leads to microstructural changes and among them, to the development of a texture induced by hot deformation of the polycrystalline alloy. This neutron diffraction texture study aims to demonstrate the {001} crystallographic fibre texture of the $Nd_2Fe_{14}B$ phase developed along the deformation direction. The starting alloy already shows a certain texture in the as-cast state due to thermal gradients on cooling. This initial texture does not really affect the specific orientation developed on forging, as soon as the deformation rate is sufficient. The comparison between forging (very high strain rate) and pressing (low strain rate) shows a higher texturation level associated to higher magnetic anisotropy when the hot deformation is slow.

Introduction

Improvements in the processes used to develop extrinsic magnetic anisotropy are needed for the engineering of high $(BH)_{max}$ magnets. Hot forging (at 1173 K, in the brittle state of $Nd_2Fe_{14}B$) has been used to produce a bulk magnet from the semi-solid Nd-Fe-B-Cu alloy. For the first time, high quality magnets (H_c =10 kOe, B_r = 1 T, BH_{max} = 24 MGOe) have been obtained in the range of very high strain rates (>100 s^{-1}) [1]. A rheological study demonstrated correlations between the permanent magnet properties and the mechanical behaviour of the Nd-Fe-B-Cu alloy during the hot deformation process [2]. Microstructural changes are the key for the two main permanent magnet properties: coercivity and macroscopic magnetic anisotropy. Coercivity results mainly from an important size reduction of magnetic $Nd_2Fe_{14}B$ crystallites and a homogeneous distribution of the intergranular phase, among other microstructural changes. The macroscopic magnetic anisotropy arises from the development of a crystallographic texture of the $Nd_2Fe_{14}B$ phase. In this study neutron texture analysis is used: 1) to demonstrate the effect of the hot deformation process, 2) to comment on the as-cast starting alloy, as far as its cooling texture is concerned and 3) to compare a rapid (forging) and a slow (pressing) deformation process.

Experimental details

Our analyses are based both on neutron texture studies and magnetic measurements. Neutron diffraction experiments have been performed on the D1B beamline (λ = 2,532 Å) at ILL, Grenoble, using the Eulerian cradle and the curved position sensitive detector. A routine procedure based on the individual fit of peaks (or groups of peaks) and data reduction was used [3]. The ODF was refined using the WIMV algorithm as implemented in Beartex [4].

Magnetic measurements provide information on the volumic magnet performances, which depend not only on the crystallographic alignment but also on the developed microstructure in terms of nature and correlations between phases present in the sample. Results of magnetic measurements showed that the direction parallel (respectively perpendicular) to the forging direction is the easy (remanence $M_r^{//}$) (respectively hard remanence $M_r^{\perp}$) magnetization direction. The macroscopic magnetic anisotropy ratio $M_r^{//}/M_r^{\perp}$ is somehow linked to the texture level in the sample. However, magnetic measurements are directional and cannot reveal the complete texture, are influenced by the crystallite shapes, and may also contain pure magnetic effects [5], but this ratio equals to 1 for isotropic properties [6]. Furthermore, it does not give in our case directly the fraction of grains oriented along the forging direction, since it depends on the initial texture of the as-cast alloy and on the forging direction with respect to this initial texture.

The $Nd_{15.5}Fe_{78}B_5Cu_{1.5}$ ingots were induction melted under argon atmosphere and cast into either cylindrical or "book" moulds (Fig. 1). They were then forged at 1173K (with a speed of 4.5 $m.s^{-1}$) or only pressed (at 0.4 $m.s^{-1}$). The height reduction of the sample is defined as h_i/h, where h_i and h are the sample heights before and after forging respectively.

The as-cast cylindrical samples (forged or pressed) have been glued on the sample holder with their casting direction parallel to the φ-axis of the goniometer (centre of pole figures), while for the book mould sample the φ-axis was perpendicular to the casting direction. Each sample had a cubic shape with 5 mm edges. A full 5° x 5° measurement grid has been performed on the book mould sample, while for the others two φ-scans (at χ=0° and χ=90°) revealed an axially symmetric texture and only a χ-scan was measured. We used two incident angles for each measurement (ω=10° and ω=30°), in order to minimise the blind area on significant pole figures.

As-Cast Textures

Book Mould Texture. This study of the as-cast book mould sample intends to confirm the stabilisation of a texture induced by thermal gradients while solidification in the mould. The thermal gradient $\vec{\nabla T}$ is perpendicular to the casting direction $\vec{M}$ (Fig. 3a). The ODF refinement

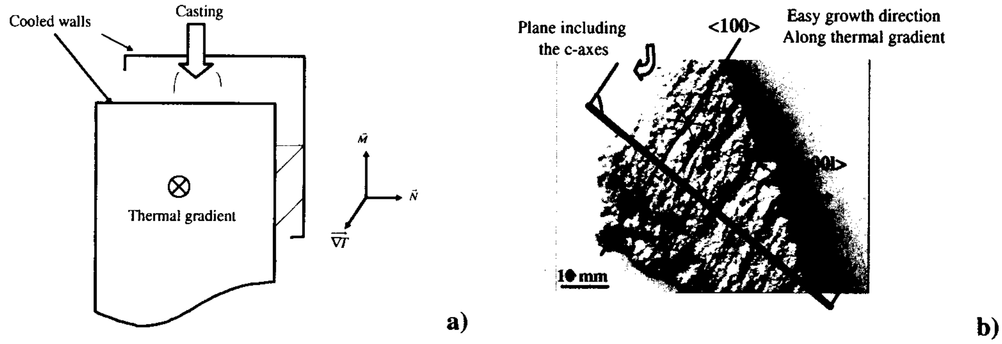


Fig. 1: a) Book mold casting and **b)** broken face and crystallographic directions

was satisfactory with RP0 = 19 % and RP1 = 16.4 %. No FON remains with a maximum ODF of 45 m.r.d. and a texture strength of F2 = 5.2 m.r.d.2. The recalculated {100} pole figure (Fig. 2) shows a marked texture with the a-axes of the Nd2Fe14B structure (P42/mnm) [6] along the thermal gradient direction, with a maximum density of 10.9 m.r.d.. The {001} pole figure reveals a nearly isotropic distribution of c-axes perpendicularly to the <100> direction and thermal gradients. This observation is in good agreement with the proposed growth mechanism for Nd2Fe14B particles as denoted in the literature [7], with thermally induced growing directions in the (a,b) planes. The deviation from axial symmetry of the {001} pole figure comes from second minor components visible in {100} as small arcs around = 45°.

Cylindrically mould Texture. An image of the sample broken perpendicularly to the casting direction shows a morphological radial alignment of crystals (Fig. 3a). A φ-rotation of the sample at

χ=0° leaves invariant diffraction peaks (Fig. 3b), pointing out that thermal gradients in the mould induce a radial growth of the crystallites with an axial symmetry around the casting direction. Very low texture levels were however detected by measurements of the χ-scans in these samples and no quantitative texture analysis was performed.

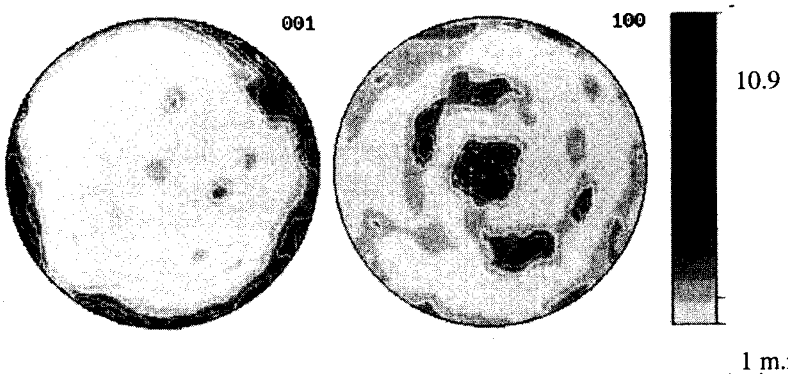


Fig. 2: {001} and {100} recalculated pole figures of the book mold sample. Equal area projection, linear density scale. $\vec{\nabla T}$ is vertical to pole figure planes, $\vec{M}$ is horizontal.

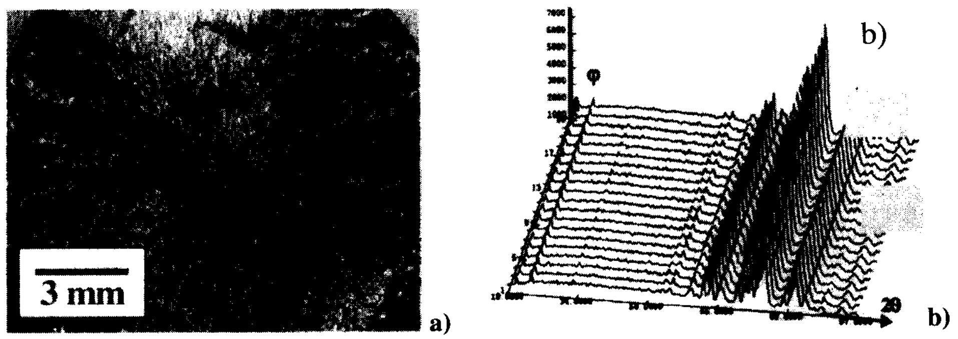


Fig. 3: Radial crystallisation texture obtained by casting in a cylindrical mould **a)** broken surface perpendicular to the casting direction and **b)** φ-scan, at χ=0°: intensities are invariant.

Texture induced by hot forging

Forging Direction Influence on Book Mould Samples. The book mould as-cast microstructure may be well suited for the development of a deformation texture during the forging step, because it already suppresses one degree of freedom for the c-axes orientations (Fig. 2). $Nd_2Fe_{14}B$ crystallites are platelet-shaped, with c-axes perpendicular to the platelet (Fig. 4) and forging an original book mould sample along its initial casting direction (Fig. 5) could result in an easiest alignment of c-axes with the casting direction than if they where initially randomly distributed. Accordingly, the highest magnetisation (higher c-axes volume fraction along the forging direction) should be obtained with the forging direction perpendicular to the thermal gradients (i.e. casting direction).

Magnetic anisotropy results on two book mould samples, hot forged in the same conditions are presented in Table 1. After forging, the magnetic orientation parameters of the two samples are nearly the same (slightly below 70%). The sample forged perpendicularly to the casting direction exhibits a magnetic anisotropy similar to the one forged parallel, thought slightly lower as a sign of an incompletely destroyed initial casting texture. The hard directions of magnetisation are not isotropic. The initial hardest direction (thermal gradient direction in the as-cast state) is maintained in the forged state.

Forging Direction Influence on cylindrically mould samples. A first φ-scan (Fig. 6) on the sample forged parallel to the casting direction confirmed the isotropic distribution of crystallographic orientations around the forging direction. In this case a fibre texture with $Nd_2Fe_{14}B$ c-axes parallel to the forging direction is developed (Fig. 7), weaker than for the forged book mould sample (only 2.4 m.r.d. at maximum of {001}).

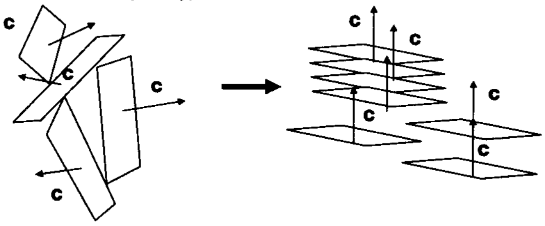


Fig. 4: Schematic effect of forging on the platelet-shaped $Nd_2Fe_{14}B$ crystallites. Initially random crystallites (left) are brought with their c-axes along the forging direction (vertical, left)

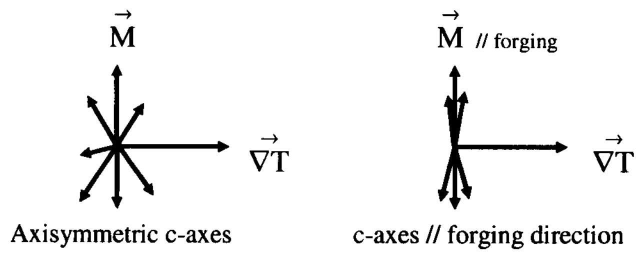


Fig. 5: Texture of a book mould sample before (left) and expected after (right) the forging process.

Forging direction	⊥ casting direction (// thermal gradient)	// casting direction
$M_r^{\perp}$ (T) (hard direction)	0.55	0.59 (⊥ thermal gradient) 0.44 (// thermal gradient)
Orientation rate ($M_r^{//}$ / M_S) (M_s : saturation magnetisation)	0.65	0.68

Table 1: Comparative anisotropic magnetic results on two book mould samples forged respectively perpendicularly and along the casting direction.

This texture is particularly adequate in terms of magnetic performances since the magnetisation vector lies along the c-axis of each $Nd_2Fe_{14}B$ crystallite. With this configuration, remanence is enhanced in the forging direction and reduced perpendicular to it. The magnetic and texture characterisations of the two studied forged samples are summarised in Table 2.

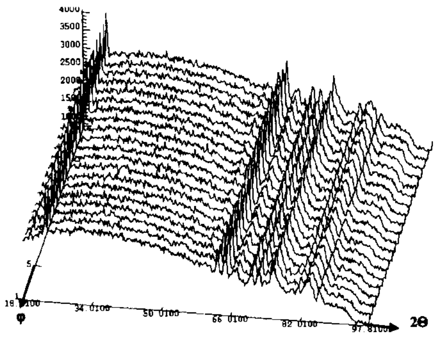


Fig. 6: φ-scan at χ =0° and ω = 10°: intensities are invariant.

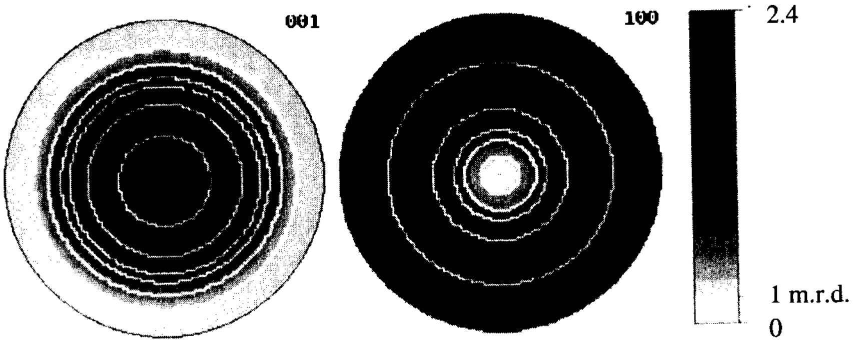


Fig. 7: {001} et {100} recalculated and symmetrised pole figures of the forged sample. Logarithmic density scale, equal area projection.

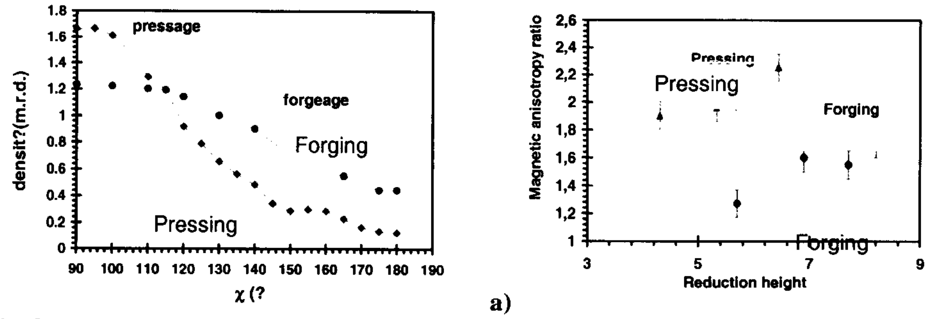
These latter are very similar for the two samples, with an even higher {001} pole density at maximum for the sample forged perpendicularly to the casting direction, this result being also visible on the magnetic anisotropy ratio. However, the reduction ratio indicates that for the same applied strain rate the sample was much deformed when the forging direction was applied perpendicularly to the casting direction. Consequently a slightly higher pole density may be attributed to this larger deformation. We conclude that in the case of a cylindrically mould sample with a low casting texture, forging direction with an enough deformation rate is not influencing much the magnetic and crystallographic anisotropies.

Forging direction	⊥ casting direction	// casting direction
Reduction ratio: h_i / h	4.4	5.8
Magnetic anisotropy ratio	1.75	1.63
{001} poles density (m.r.d.)	2.6	2.4

Table 2: Comparative results on two cylindrically mould samples forged respectively perpendicularly and along the casting direction.

Forged versus Pressed Textures

Two cylindrically mould samples have been pressed (0.4 m.s-1) and forged (4.5 m.s-1) parallel to the casting direction with a height reduction of 6.5 in order to reveal eventual effects of strain rates during hot deformation. The measured magnetic anisotropy ratio are 2.3 and 1.6 respectively. For the two samples we observe a <001> fibre texture (Fig. 8a) with the fibre axis along the casting direction. The pressed sample shows a more pronounced crystallographic texture than the forged one with 1.7 m.r.d. and 1.2 m.r.d. at maximum of the distribution respectively, coherently with the decrease of the magnetic anisotropy ratio. This ratio slightly increases with the deformation (Fig. 8b) but remains always higher at low strain rates (pressed sample).



a) b)

Fig. 8: a): {002} recalculated pole density evolution versus χ for the forged and pressed samples and b): Magnetic anisotropy ratio versus height reduction for pressed and forged samples.

Conclusion

Whatever the casting texture, the forging process of the as-cast alloy enables to dominate the subsequent orientation of the $Nd_2Fe_{14}B$ phase, with c-axes along the forging direction being a fibre axis. Starting with a stronger initial texture, i.e. with the help of a thermal gradient as in the case of a book mould sample allows to achieve stronger final texture after forging in the proper direction. However, the casting texture is not a critical parameter in cylindrically mould samples for the development of extrinsic magnetic anisotropy by the hot deformation process, as soon as the deformation is large enough and the strain rate slow enough to allow the texture to develop. In this sense, the comparison between pressed and forged cylindrically mould samples indicates a more pronounced texture effect for a slower deformation process (pressing).

Acknowledgements

This work has been funded by the European Union project under the Growth program (G6RD-CT99-00169) ESQUI: "X-ray Expert System for Electronic Films Quality Improvement".

References

[1] S. Rivoirard, P. de Rango, D. Fruchart, R. Perrier de la Bâthie: patent n° 9806745 (1998).

[2] S. Rivoirard, P. de Rango, D. Fruchart, Y. Chastel, C.L. Martin: Mater. Sci. and Engineering A311 (2001), 121.

[3] D. Chateigner, H.R. Wenk, M. Pernet: J. Appl. Cryst. 30 (1997), 43.

[4] H.R. Wenk, S. Matthies, J. Donovan, D. Chateigner: J. Appl. Cryst. 31 (1998), 262.

[5] M. Morales, D. Chateigner, D. Fruchart: To be published J. Magn. Magn. Mat..

[6] W. Heisz, L. Schulz: Appl. Phys. Lett. 53 (1988), 342.

[7] J.F. Herbst, J.J. Croat, F. E. Pinkerton: Phys. Rev. B 29 (7) (1984), 4176.

[8] P. Tenaud, A. Chamberaid, V. Vanoni: Solid State Comm. 63 (1987), 303.