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Texture development in Nd-Fe-V alloys by hot deformation in view of permanent magnet properties

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Abstract.

In this study, a hot forging process is applied to Nd-Fe-V as cast alloys in order to develop both the microstructure and the crystallographic texture appropriate for permanent magnet properties. A neutron diffraction texture analysis is used to account for the extrinsic magnetic anisotropy: the stabilisation of the $Nd(Fe,V)_{12}$ hard magnetic phase has been achieved during forging but its extrinsic anisotropy level remains low. Attempts to understand this phenomenon are made through a discussion on the Nd-Fe-V alloy rheological and mechanical behaviour and a comparison with Nd-Fe-B permanent magnets.

Introduction

Good levels of coercivity and enhanced magnetic anisotropy are the key for high performance magnets. In the field of rare earth - transition metal alloys, the development of anisotropic powders is needed for bonded magnet applications. Anisotropic Nd-Fe-B based alloys are already commercially available whereas, only recently some studies report on Nd-Fe-V-N alloys with potential permanent magnet properties.

The $NdFe_{10.5}V_{1.5}$ compounds, one of the 1:12 phases $RFe_{12-X}M_X$ (R = rare earth, M = transition metal), can be considered as potential materials for permanent bonded magnets because of their high intrinsic magnetic properties, higher than those of the $Nd_2Fe_{14}B$-type materials [1], which are presently the most used as permanent magnets. For this purpose, good extrinsic magnetic properties are also needed: coercivity, and extrinsic magnetic anisotropy. Up to now, mechanical alloying enabled to obtain a high coercivity in this type of materials (H_c = 872.17 kA/m) [2].

However, large-scale production processes have to be considered. We proposed to apply a high-speed hot forging process to induce the microstructure suitable for permanent magnet properties in Nd-Fe-V alloys. In the case of Nd-Fe-B materials, coercivity and extrinsic anisotropy were already successfully induced in the bulk alloy by applying this hot forging process with a strain rate of 125 s^{-1}[3]. Coercivity resulted from grain size reduction. Extrinsic magnetic anisotropy was linked to a fibre texture of the c-axes, being the easy axes of magnetisation [4].

Besides the extrinsic magnetic properties, the major concern in the preparation of Nd-Fe-V alloys is the stabilisation of the $NdFe_{10.5}V_{1.5}$ phase. The compounds can contain free iron, detrimental to the coercivity and the temperature and composition range of the 1:12 phase stability domain is limited. The high-speed hot forging process applied to as-cast Nd-Fe-V alloys, (containing only free iron and an intergranular Nd-rich phase) allowed the stabilisation of the 1:12 phase and the almost complete dissolution of free iron [1]. This revealed equivalent to a long annealing treatment of 24 hours at 960°C. The two main rheological parameters of the forging process were optimised, (the temperature and the Nd content). The best results were obtained by forging at 930°C samples prepared with 10% Nd-excess.

Experimental details

Samples of the $NdFe_{10.5}V_{1.5}$ composition with 10% excess of Nd were prepared by induction melting and then cast into cylindrical stainless steel tubes of 12 mm in diameter. The forging process was applied on these ingots under argon atmosphere at 930°C, either in the as-cast state, presenting free iron dendrites and the Nd-rich intergranular phase, or after an annealing treatment for 24 hours at 960°C, leading to the homogeneous 1:12 phase, without free iron traces. A third sample, also annealed, was prepared with 3% Nd-excess, in order to determine the influence of the intergranular phase content on the texture. At the forging temperature, the intergranular phase is liquid whereas the 1:12 phase or the free iron dendrites are solid.

The texture of the $NdFe_{10.5}V_{1.5}$ alloys after the high-speed hot forging process was studied by neutron diffraction on the D20 powder diffractometer of the Laue-Langevin high flux nuclear reactor in Grenoble, France. For this purpose, an Eulerian circle was mounted on the diffractometer. The multidetector registers all the pole figures simultaneously for each (χ,φ)-position.

The samples, of cubic shape of about 6mm, were put on the Eularian circle in such a way that the forging direction was in a plane perpendicular to the φ-axis. The position of the circle was $\omega = 34.2°$ corresponding to the Bragg angle of the (400) peak of the 1:12 structure. Scans in χ from 0° to 90° with a 5° step and in φ from 0° to 355° with a 5° step for each value of χ were operated for all the analysed samples.

Data treatment was carried out using the following calculation codes: XRFIT (developed at ILL), POFINT [5], GOMAN [6] and Beartex [7].

Results

For all the samples, a fibre texture component is observed, with the <010> directions parallel to the forging direction Y, reinforced by two three-dimensional components. This is presented in Figure 1, in the case of the sample with 10% Nd-excess. The {100} and {001} pole figures illustrate the presence of a nearly fibre texture with <010> // Y being the fibre axis. These figures also evidence two texture components with c-axes at about $\chi = 60°$.

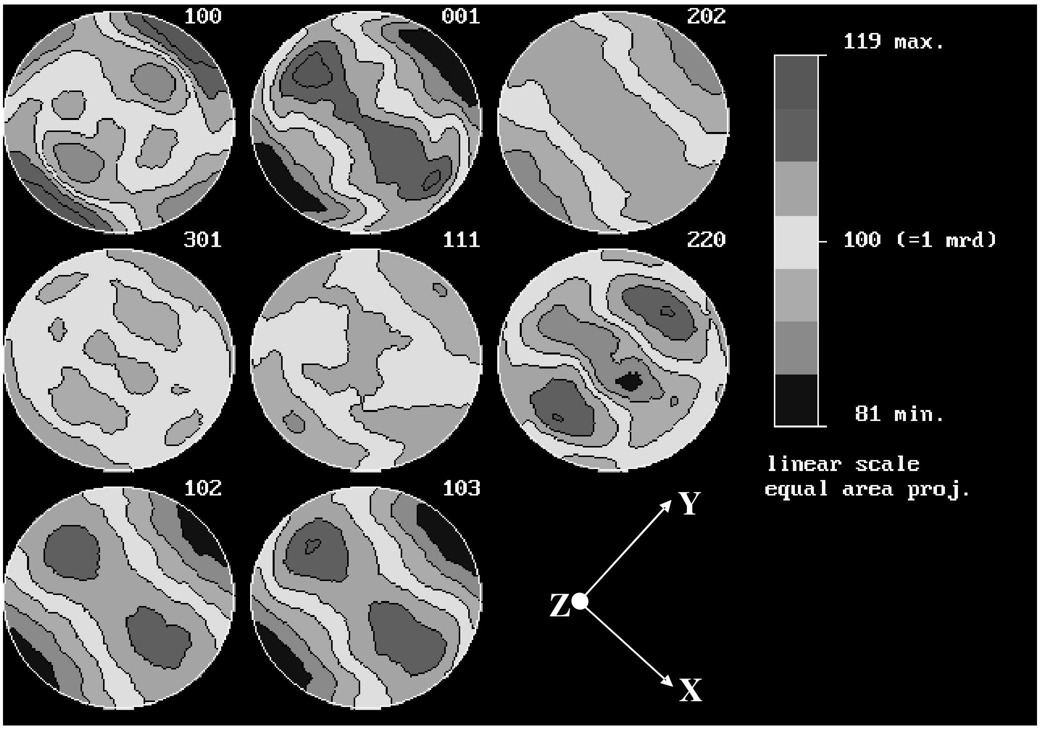

Figure 1. Low (hkl) poles figures recalculated from the ODF for $NdFe_{10.5}V_{1.5}$ + 10%Nd

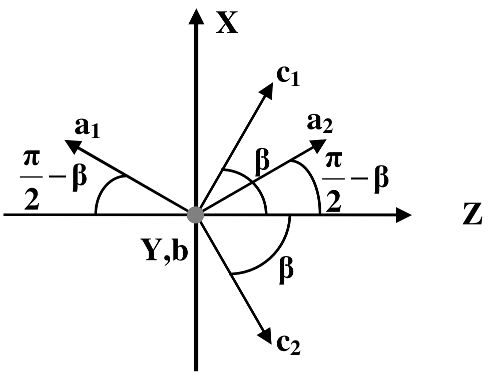

Figure 2. Three-dimensional texture components obtained after forging the $NdFe_{10.5}V_{1.5}$ + 10%Nd sample. Y is the forging direction and (a_1, b_1, c_1), (a_2, b_2, c_2) the two texture components

Using the Roe-Matthies convention [8, 9], the two three-dimensional components are defined by {0,β,0} and {π,β,0} (β ≈ 60°).

Results obtained on the three studied samples (Table 1) show that 36% to 71% of the material remains untextured after process, with correspondingly low texture strengths, strongest textures being achieved after annealing and with less Nd excess. Reliability of the ODF refinements are satisfactory.

Sample	**ODF min** (mrd)	**ODF max** (mrd)	**F^2** (mrd^2)	**RP0** (%)	**RP1** (%)
3%Nd excess, annealed	**0.35**	**2.40**	**1.0297**	**1.4133**	**1.3773**
10%Nd excess, annealed	**0.62**	**1.47**	**1.0134**	**2.2880**	**2.2123**
10%Nd excess, as-cast	**0.71**	**1.31**	**1.0044**	**1.1960**	**1.1422**

Table 1. Texture results for the different $NdFe_{10.5}V_{1.5}$ samples, forged at 930°C.

The nearly fibre texture is in good agreement with the axial symmetry of the deformation process. However, since for $NdFe_{10.5}V_{1.5}$ the easy axis of magnetisation is the c-axis, the b-axis fibre texture developed along the forging direction is not the one that favours development of extrinsic magnetic anisotropy. The distribution of the easy magnetisation c-axes quite at random in a plane perpendicular to the forging direction, even if slightly reinforced along the texture components is not suitable for the macroscopic use of the intrinsic magnetic anisotropy. Moreover, the high volume fraction of randomly oriented grains is also detrimental to macroscopic magnetic anisotropy. This result can explain the typical isotropic magnetic behaviour observed on these samples, with a remanence of about one half of the saturation magnetisation. Furthermore, the intergranular phase content influences texture development: with 3 to 10% of Nd-excess this detrimental randomly oriented volume fraction increases from 36% to 62%.

A comparison between the two samples containing 10%Nd-excess proves the influence of the initial microstructure on the texture induced by the forging process. For the sample forged in the as-cast state, 71% of the 1:12 phase is randomly oriented, while it reaches only 62% for the sample annealed before forging.

Discussion

For Nd-Fe-V alloys, small intergranular phase contents (typically <5% in volume) do not allow solid grains orientation in the liquid intergranular phase at the forging temperature. This rheological behaviour is however different from the one of Nd-Fe-B alloys for which the deformation mechanism is mainly related to an homogeneous lubricated flow in the semi-solid state

[3]. The alloy microstructure observed after forging (Figure 3), rather suggests that the main mechanical behaviour is due to a solid state deformation process. Indeed, scanning electron microscopy studies on the forged samples reveal, inside the crystallites, white parallel lines which are neither coming from chemical composition nor magnetic contrasts (Figure 3a,b). The observation of this contrast in secondary electrons mode only suggests a crystallographic or topographic origin and supports the existence of sliding planes in the $NdFe_{10.5}V_{1.5}$ solid phase. This type of microstructure is an indication of the irreversible stress and strain, taking place between 1:12 adjacent grains inside the alloy during the forging process. Brittle behaviour of the 1:12 phase is observed in some regions (Figure 3c) where the strains are concentrated.

Activation of sliding planes under the effect of uniaxial load must lead to an orientation of these planes perpendicular to the forging direction. Looking at texture results indicate that the planes perpendicular to Y are the (0k0) planes, which are dense planes in this structure, and then are favoured for crystal sliding under forging conditions.

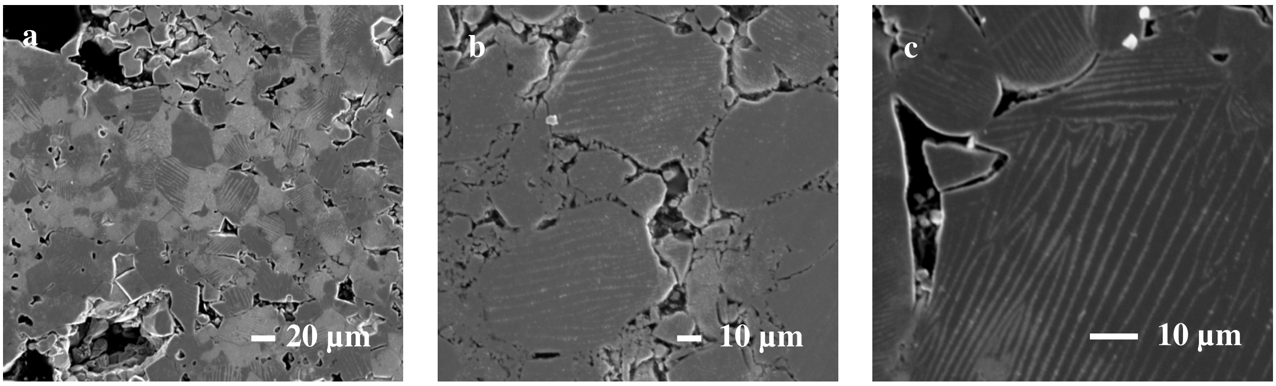

Figure 3. $NdFe_{10.5}V_{1.5}$ alloys +3% Nd microstructure after forging at 930°C

Nevertheless, the randomly oriented volume differences observed between the 3 and 10% of Nd samples suggest that the influence of the intergranular liquid phase cannot be neglected. The 1:12 phase solid crystallites do not present any shape anisotropy, thus a random orientation of the crystallites due to a liquid flow could be expected as a minor additional deformation mechanism. Depending on its amount, the liquid phase could act as a lubricant to accommodate intergranular crystallite deformation or even enable the crystallite rotation after sliding. In this case, part of the imposed forging stress is relaxed and the crystallographic texture is less developed.

Another mechanism can be identified in the forged samples. The 1:12 phase crystallisation occurs during the forging process from free iron and the Nd-rich intergranular phase. This crystallisation adds to the deformation mechanism, and newly formed crystals are subjected to both uniaxial orientation process and grain contacts. This results in roughly the same texture as in other samples, except that it may explain the three dimensional texture components developed. These components are then more developed for the annealed samples where crystallisation is enhanced. However, since crystallisation speed anisotropy is weak (crystallites are isotropic), such crystallisation does not promote a strong texture by grain reorientation under pressure, and then accounts for high volume fraction of randomly oriented grains.

The three dimensional texture components could also be provided by the combination of secondarily active sliding planes (like (hk0) planes) and another pressure mechanism perpendicular to Y. Such a transverse pressure may occur from material flows during the strong deformation.

Conclusion

Neutron diffraction studies of $NdFe_{10.5}V_{1.5}$ forged alloys reveal a fibre texture associated to two three-dimensional texture components. The fibre texture is induced by sliding of (0k0) planes under the mechanical strain. The <001> directions of the two three-dimensional components make an angle of 60° with the Z-axis. This texture is not suitable for extrinsic magnetic anisotropy and permanent magnets properties development.

The same texture types are observed for an as-cast alloy and for an alloy annealed before forging. Influences of initial microstructure, intergranular phase content and isotropic crystal growth all are partly removing orientation influence of the driving force and create high levels of randomly oriented material.

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