A NOVEL PROCESS FOR RARE EARTH-IRON-BORON PERMANENT MAGNETS PREPARATION

J. Nozières, R. Perrier de La Bathie, J. Gavinet

JOURNAL DE PHYSIQUE
Colloque C8, Supplément au n° 12, Tome 49, décembre 1988

A NOVEL PROCESS FOR RARE EARTH-IRON-BORON PERMANENT MAGNETS PREPARATION

J. P. Nozières (1), R. Perrier de la Bathie (1) and J. Gavinet (2)

(1) *C.N.R.S., Laboratoire Louis Néel, 166 X, 38042 Grenoble Cedex, France*
(2) *L.E.T.R.A.M. C.E.N., Saclay, 91191 Gif-sur-Yvette Cedex, France*

Abstract. – A novel process for permanent magnets fabrication is described: low cost and good energy products are achieved by hot-working cast ingot of NdFeB alloys. Magnetic characterization and microstructural observation reveal that the coercivity mechanisms are essentially similar to those observed in sintered magnets.

Introduction

It is shown in this paper that large coercivity can be directly obtained by hot working cast ingots of NdFeB Alloys. A brief description of the preparation process is given, the coercivity mechanisms are discussed and related to the observed microstructure.

Process description [1]

Alloy is prepared by induction melting of commercial products. The ingot composition is approximately $Nd_{15.5}Fe_{78}B_6Al_{0.5}$ at. % which corresponds to the one used for preparation of magnets by the powder metallurgy method. To improve magnetic properties, additives such as Dy or Co may be used. The as-cast ingots are hot-worked under controlled atmosphere or after sealing in an iron sheath. Different types of hot working processes can be applied: extrusion, hot-pressing, rolling, hammering. The hot-working temperature ranges between 750 °C and 950 °C with little influence on the magnetic properties. At such temperatures, the intergranular phases which represents about 10 % of the total sample volume, are liquid. Thanks to the coexistence of a liquid and a solid phase, the matter will keep homogeneous during the welding (i.e. a mechanical heat treatment involving da plastic deformation). High welding rates are necessary to provide the optimal grain size and a partial magnetic orientation; they vary from 5 to 25. A final heat treatment is applied to improve and stabilize the magnetic properties. No general rule has been found yet. However, melting of the intergranular phases as well as defect relaxation are expected to play an important role.

Microstructure observations

Primary crystallization is of little importance on the final permanent magnet properties. A 50-100 μm structure is obtained with normal cooling conditions ($\sim 300 - 400°$/mn). Mechanical treatment breaks the grains below the critical size of 10 to 15 μm where coercivity appears. The microstructure is inhomogeneous, few unbroken grains around 20 μm are disseminated in a matrix made of grains with typical size 0.5 to 10 μm. No dynamic recrystallization has been observed. The same phases as in sintered magnets are observed [2] i.e. the $Nd_2Fe_{14}B$ main phase, the boron rich phase $Nd_{1+\varepsilon}Fe_4B_4$ and an intergranular phase made out of low melting Nd-rich eutectics involving Nd solid solution, NdAl eutectics, oxydes... Differential thermal analysis reveals for these intergranular phases melting points in the vicindity of 700 °C. A soft phase which is assumed to be free iron was also observed.

After annealing, a slight increase of the smallest grains is noticed up to about 1 μm and the grains tend to have a rounder shape. The iron-rich phase is no more observed in the annealed samples.

Preferential orientation of grains

It is observed that preferential orientation of grains is obtained directly by mechanical hot-working during which the c-axes are eliminated from the hot-working direction. Starting from totally isotropic ingots, all the easy-axes are lying in the plane perpendicular to the hot-working direction. It is possible to improve the preferential orientation by using directed primary crystallization. As the growing direction of the $Nd_2Fe_{14}B$ phase is [100], the c-axis of the cast-ingots are distributed in the plane perpendicular to the direction of the thermal gradient applied during cooling. If the hot-working direction is subsequently applied in the plane containing the c-axes, a unique axis is in principle selected. At the present stage, only a partial orientation of grains is obtained, leading to values of the remanent induction around 9 kG.

Magnetic properties

Typical demagnetization curves at 300 K are shown in figure 1. The coercive field is of about 11 kOe, $(BH)_{max}$ of 15 MGOe. Values of 20 MGOe have already been reached.

In order to analyse the coercivity in these magnets, first magnetization curves were measured. A high re-

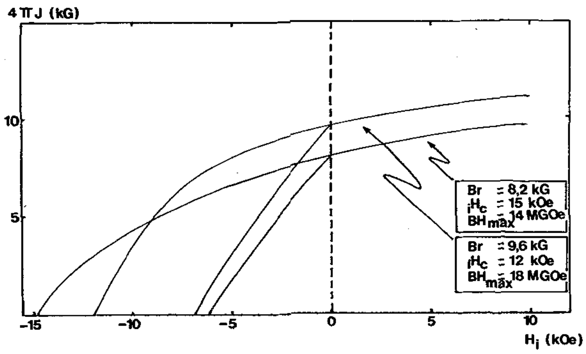

Fig. 1. – Typical demagnetization curve at 300 K.

versible susceptibility is observed revealing that domain walls movre freely. No singnificant pinning is observed, even before defect relaxation by heat treatment. Coercivity develops as the saturation field is increased from about 5 to 25 kOe. For a given value of the saturation field, it appears that grains in the magnet either exhibit full coercivity, close to the macroscopic coercive field of the magnet, or do not have any significant coercivity. As the saturation field is increased, the percentage of coercive grains is also increased. In a given saturation field, this percentage is almost identical at 300 K and 175 K. All the above observations have recently been observed in NdFeB sintered magnets [3]. This strongly suggests that the coercivity mechanisms in magnets prepared by the novel process described here are the same as these involved in NdFeB sintered magnets.

The temperature dependence of the coercive field from 4.2 K to 400 K is shown in figure 2. The coercive field at 4.2 K is 48 kOe, and vanishes at about 500 K. This variation was fitted by the relation:

$$H_0 = H_c + 25S_v = \alpha\frac{\gamma v^{-1/3}}{M_s} - \beta\, 4\pi M_s \qquad (1)$$

which has been previously proposed for sintered magnets [4]. In relation (1), S_v is the viscosity coefficient, γ is the domain wall energy per unit area, v the value of the activation volume, M_s the spontaneous magne-

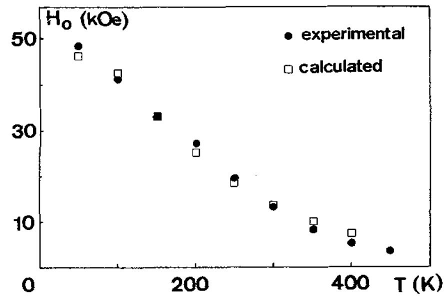

Fig. 2. – Temperature dependence of the coercive field.

tization of $Nd_2Fe_{14}B$ phase. The best fit was obtained for values of the free parameter (see Fig. 2): $\alpha = 0.35$ and $\beta = 0.32$.

Conclusion

A novel Rare Earth-Iron-Boron permanent magnet production process has been developed. It combines low cost, simplicity and sufficient magnetic properties for many industrial applications, particularly for motor devices. Thanks to the hot working process any type of profiles can be obtained in large quantities. An analysis reveals that the microstructure is very inhomogeneous and contains essentially the same phases as the sintered magnets. The magnetic properties suggest that coercivity mechanisms are the same as in sintered magnets.

[1] Perrier de la Bathie, R., Chavanne, J., Brevet PCT/FR 87/00175.

[2] Schneider, G., Henig, E. T., Petzow, G., Stadelmaier, H. H., *Z. Metallkd Pd* **77** (1986) H 11, p. 755.

[3] Givord, D., Lu, Q., Tenaud, P., Viadieu, T., unpublished;
Tenaud, P., Thèse Univ. J. Fourier, Grenoble.

[4] Givord, D., Tenaud, P., Viadieu, T., *I.E.E.E. Trans. Magn. Mag.* **24** (1988) 1921.