

M. DOŚPIAŁ<sup>\*,#</sup>, M. NABIAŁEK<sup>\*</sup>, M. SZOTA<sup>\*\*</sup>, J. GONDRO<sup>\*</sup>

## THE MAGNETIZATION REVERSAL PROCESSES OF BULK (Nd, Y)-(Fe, Co)-B ALLOY IN THE AS-QUENCHED STATE

### PROCESY PRZEMAGNESOWANIA MASYWNYCH STOPÓW (Nd, Y)-(Fe, Co)-B W STANIE PO ZESTALENIU

The magnetization reversal processes of bulk  $\text{Fe}_{64}\text{Co}_5\text{Nd}_6\text{Y}_6\text{B}_{19}$  alloy in the as-quenched state have been investigated. From the analysis of the initial magnetization curve and differential susceptibility versus an internal magnetic field it was deduced, that the main mechanism of magnetization reversal process is the pinning of domain walls at the grain's boundaries of the  $\text{Nd}_2\text{Fe}_{14}\text{B}$  phase. Basing on the dependence of the reversible magnetization component as a function of magnetic field it was found that reversible rotation of a magnetic moment vector and motion of domain walls in multi-domain grains result in high initial values of the reversible component. The presence of at least two maxima on differential susceptibility of irreversible magnetization component in function of magnetic field imply existence of few pinning sites of domain walls in  $\text{Fe}_{64}\text{Co}_5\text{Nd}_6\text{Y}_6\text{B}_{19}$  alloy. The dominant interactions between particles have been determined on the basis of the Wohlfarth dependence. Such a behavior of Wohlfarth's plot implies that the dominant interaction between grains becomes short range exchange interactions.

*Keywords:* magnetization reversal, hard magnetic materials, nano-materials

W pracy przedstawiono wyniki badań własności magnetycznych i mechanizmów przemagnesowania masywnego stopu  $\text{Fe}_{64}\text{Co}_5\text{Nd}_6\text{Y}_6\text{B}_{19}$  w stanie po zestaleniu. Z krzywych pierwotnego namagnesowania i ich podatności różnicowej określono, że głównym mechanizmem przemagnesowania jest kotwiczenie ścian domenowych. Na podstawie zależności składowych odwracalnych namagnesowania od pola magnetycznego stwierdzono, że procesy odwracalne takie jak obroty wektora namagnesowania oraz ruch ścian domenowych w wielodomenowych ziarnach mają znaczny wpływ na zmiany namagnesowania dla początkowych wartości pól magnetycznych. Obecność maksimum w zależności składowej nieodwracalnej podatności magnetycznej świadczy, że głównym mechanizmem przemagnesowania w badanych magnesach jest kotwiczenie ścian domenowych na licznych centrach kotwiczenia znajdujących się na granicach ziaren fazy  $\text{Nd}_2\text{Fe}_{14}\text{B}$  oraz na defektach strukturalnych. W celu określenia oddziaływań między ziarnami określono zależność Wohlfartha. Zależność ta nie jest spełniona dla badanego magnesu, świadczy to o istnieniu oddziaływania wymiennego między ziarnami magnesu.

### 1. Introduction

The investigated  $\text{Fe}_{64}\text{Co}_5\text{Nd}_6\text{Y}_6\text{B}_{19}$  alloy was obtained using suction-casting method, in which the melted material is sucked into a water-cooled copper mould. This method belongs to the so-called rapid cooling methods. Due to the much lower cooling rate ( $10^1$ - $10^2$  K/s) compared to classical methods (melt-spinning  $10^4$ - $10^6$  K/s) it is much harder to obtain alloy, which exhibit a good hard magnetic properties in as-quenched state [1]. Despite these complications this method has significant advantages such as: the possibility of obtaining the finished magnets without the need for further processing (eg, sintering, pressing or bonding magnetic powders); the solid material have much more better mechanical properties than the magnets obtained in the different routes; a high density has a large influence on the good hard magnetic properties. These advantages suggest

that it is possible to apply this method in the industry, for example in production of the micro engines.

The aim of this study is to determine magnetization reversal mechanisms occurring in the  $\text{Fe}_{64}\text{Co}_5\text{Nd}_6\text{Y}_6\text{B}_{19}$  as-quenched magnet, in the form of plates, obtained by suction-casting method.

### 2. Materials and methods

The ingots used in production of permanent magnet were melted using plasma arc, under a protective argon atmosphere. The elements used in production were of high purity: Fe - 99.98, Co - 99.98; Y - 99.99; Nd - 99.99. Boron was added to the alloy as a FeB ingot with known composition. Ingots were re-melted several times in order to obtain homogenous material. The sample of bulk  $\text{Fe}_{64}\text{Co}_5\text{Nd}_6\text{Y}_6\text{B}_{19}$  alloy in the

\* CZESTOCHOWA UNIVERSITY OF TECHNOLOGY, INSTITUTE OF PHYSICS, 19 ARMII KRAJOWEJ AV., 42-200 CZESTOCHOWA, POLAND

\*\* CZESTOCHOWA UNIVERSITY OF TECHNOLOGY, INSTITUTE OF MATERIALS SCIENCE, 19 ARMII KRAJOWEJ AV., 42-200 CZESTOCHOWA, POLAND

# Corresponding author: mdozial@wp.pl

as-quenched state was produced in the form of plate using a suction-casting method.

The magnetic measurements: the initial magnetization curves, the hysteresis loop, the sets of recoil curves from different points on the initial magnetization and demagnetization curves were performed by the LakeShore vibrating sample magnetometer with a maximum applied magnetic field of 2T. From the hysteresis loop and the initial magnetization curve the magnetic parameters were derived. From the recoil loops the field dependence of total magnetization  $M_{tot}$  as well as the reversible  $M_{rev}$  and irreversible  $M_{irr}$  magnetization components in both polarization directions of applied magnetic field ( $r$  – magnetization,  $d$  - demagnetization) were derived by the method described in the literature [2-4]. By differentiating the  $M_{rev}(H_i)$  and  $M_{irr}(H_i)$  the dependencies the total  $\chi_{tot}$ , the reversible  $\chi_{rev}$  and irreversible  $\chi_{irr}$  susceptibilities as a function of an internal magnetic field were obtained. The field dependencies of magnetization and their differential susceptibilities were used to determine magnetization reversal process.

The interactions between grains were examined using Wohlfarth relation:

$$\frac{M_{irr}^d(H_i)}{M_R} = 1 - 2 \cdot \frac{M_{irr}^r(H_i)}{M_R} \quad (1)$$

where  $M_{irr}^{r,d}$ ,  $M_R$ ,  $M_{irr}^{r,d}$  is the value of the magnetization when the magnetizing field is reduced to zero along the recoil curve,  $M_R$  is the magnetization when the reversed field applied to previously saturated sample is reduced to zero). Wohlfarth showed, that relationship (1) is valid for non-interacting, uniaxial, single-domain particles [5].

All magnetic measurements were taken at room temperature (21°C) for powdered samples.

### 3. Results and discussion

Fig. 1a and 1b shows the initial magnetization curve and hysteresis loop, respectively. From these curves the magnetic parameters i.e. remanence  $\mu_0 M_R = 0.59$  T, coercivity  $H_{c1} = 0.47$  T and saturation of the magnetization  $\mu_0 M_S = 0.96$  T were determined.

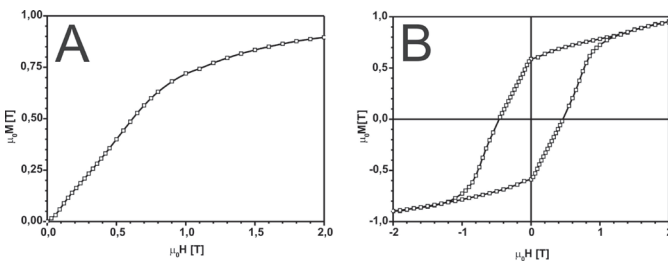


Fig. 1 Initial magnetization curve (a) and the major hysteresis loop (b) for the  $Fe_{64}Co_5Nd_6Y_6B_{19}$  sample

The initial magnetization curve shows a steep rise as a result of a combination of two types of magnetization processes. For this sample, apart from the pinning of domain

walls in hard magnetic Nd-Fe-B grains, the rotation of the magnetization vector in soft magnetic grains exists and plays the dominant role in magnetization. The non-zero slope of the initial magnetization curves at low fields indicates that another mechanism is taking part yet in magnetization process.

More details about magnetization processes can be obtained by studying the recoil loops presented in Fig. 2.

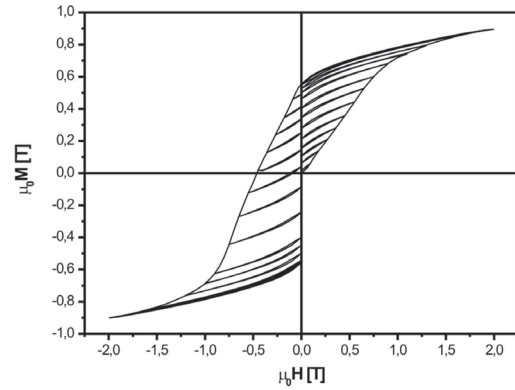


Fig. 2 Recoil curve measured in both magnetization directions for the  $Fe_{64}Co_5Nd_6Y_6B_{19}$  sample

From these curves the field dependence of total magnetization  $M_{tot}$  as well as the reversible  $M_{rev}$  and irreversible  $M_{irr}$  magnetization components in both polarization directions (Fig. 3a – magnetization, Fig. 3b - demagnetization) of an applied magnetic were derived.

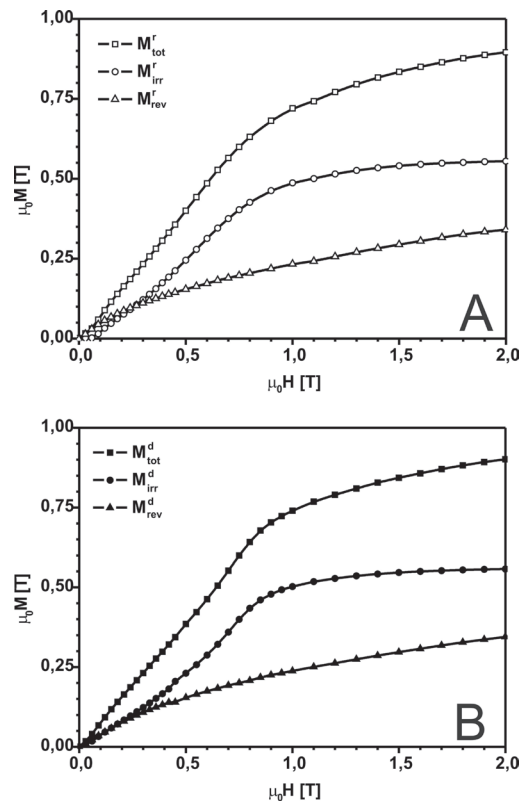


Fig. 3 Reversible  $\mu_0 M_{rev}$  and irreversible  $\mu_0 M_{irr}$  components of the total magnetization  $\mu_0 M$  during the initial magnetization (a) and the demagnetization (b) processes as a function of an internal field  $\mu_0 H_i$

In both directions of the magnetization we observe firstly rapid (up to  $\mu_0 H$  of 0.2 T) and then slow growth of a reversible component of magnetization. The field dependence of the irreversible component is more complicated. For curves measured in both magnetization directions the presence of two inflection points can be distinguished. In the field ranges from 0.1 to 1.0 and from 0 to 0.85 T measured respectively in the magnetization and demagnetization direction, a gradual increase in irreversible magnetization is observed. Above these values of the magnetic field the irreversible magnetization begins to saturate. Irreversible component measured in the magnetization direction is close to zero for the value of the magnetic field about 0.1 T. Such a contribution of the reversible and irreversible components suggest very complex reversal magnetization process [6].

In order to obtain more details about reversal magnetization,  $\chi_{irr}^r$  ( $\chi_{irr}^d$ ) reversible and  $\chi_{irr}^r$  ( $\chi_{irr}^d$ ) irreversible susceptibilities as a function of magnetic field were determined. These dependencies are shown in Fig. 4a (magnetization direction) and 4b (demagnetization direction). On both figures the total susceptibilities  $\chi_{tot}^{r,d} = \chi_{rev}^{r,d} + \chi_{irr}^{r,d}$  derived for both magnetization directions are also presented.

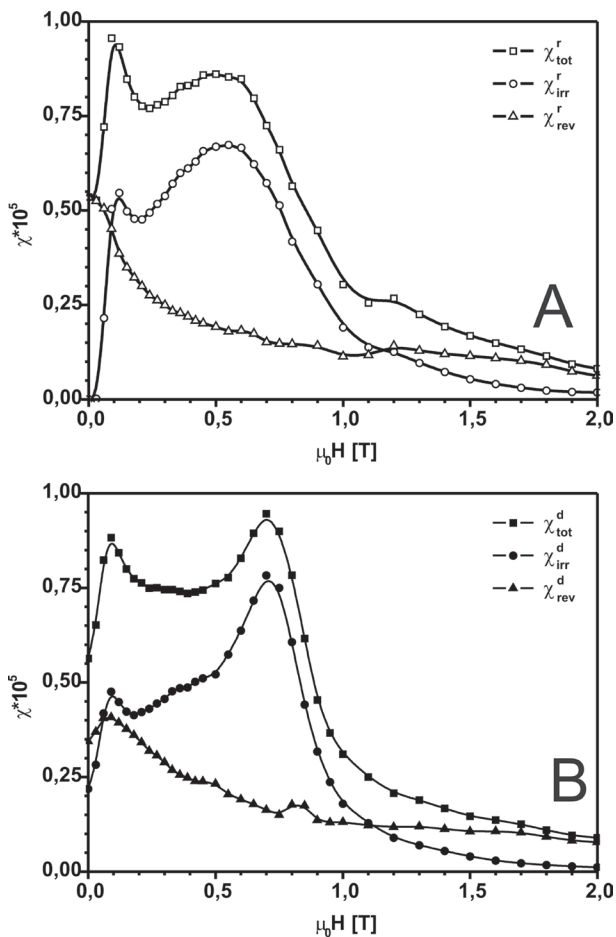


Fig. 4 Reversible  $\chi_{rev}$  and irreversible  $\chi_{irr}$  components of a total susceptibility during the initial magnetization (a) and the demagnetization (b) processes as a function of an internal field  $\mu_0 H_i$  derived for the  $Fe_{64}Co_5Nd_6Y_6B_{19}$  sample

The presence of three explicated maximums on the irreversible component of magnetic susceptibility for both magnetization directions confirms that the reversal

magnetization mechanism is dominated by pinning of domain walls at grains boundaries of hard magnetic  $Nd_2Fe_{14}B$  phase and structural defects which raised during the rapid solidification process. The coercivity of the investigated magnet is thus the sum of the contributions of the three pinning centers observed for field values equal respectively: 0.2 T, 0.35 T and 0.75 T.

According to the Wohlfarth theory [3], irreversible susceptibility meets the relation  $\chi_{irr}^d = 2\chi_{irr}^r$  (2) and a half-width of the maxima and maxima positions are the same for the magnetization and demagnetization direction [7]. A comparison of the irreversible susceptibility components in both magnetization directions testifies that this dependency is not meet. This indicates the existence of interactions between the magnet particles.

Large initial value, followed by rapid decrease of reversible susceptibility indicate a significant influence of reversible magnetization processes such as a rotation of the magnetization vector in soft magnetic phase and the motion of domain walls in multi-domain grains. The presence of small maxima on reversible susceptibility component, that are more visible in the demagnetization direction (for the values of external magnetic field near the values where the maximums on the irreversible susceptibility component are present) can be related with motion of unpinned and bowing of strongly pinned domain walls.

Fig. 5 shows the remanence curve of the investigated magnet. Wohlfarth's dependence (1) is not meet. The experimental points lie above the line for almost all except the negative values in the range from about -0.5 to -1.0 T. This proves the existence of short range exchange interactions between grains.

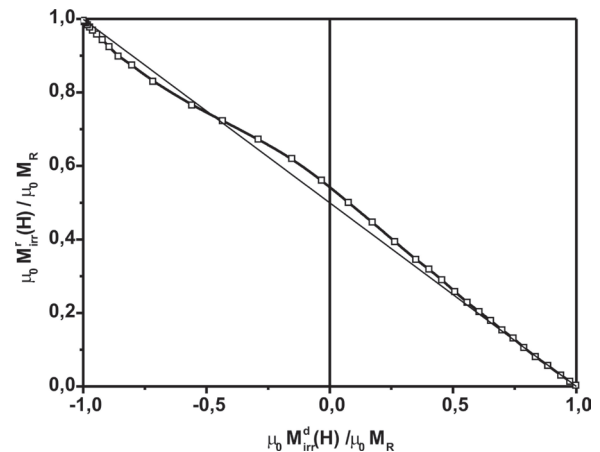


Fig. 5 Wohlfarth relationship determined from the recoil curves for  $Fe_{64}Co_5Nd_6Y_6B_{19}$  sample

#### 4. Conclusions

The suction casting method allows to produce bulk materials with good hard magnetic properties.

Produced magnet have multiphase composition, due to the overstoichiometric Fe and B addition [8]. About multiphase structure can also testify large initial value of reversible susceptibility component, resulting from presence

of soft magnetic phase and presence of several maximums at irreversible susceptibility curves, resulting from hard magnetic phases.

Analysis of reversible and irreversible components allowed to determine dominant reversal magnetization process. It was found that the main reversal magnetization mechanism is pinning of domain walls at the grain boundaries and structural defects.

As was shown by Wohlfarth et al [3] in magnets without exchange interaction between grains, remanence does not exceed half the value of saturation of the magnetization. In the investigated magnet this ratio is slightly higher ( $0.59 = \mu_0 M_R > 0.5 \mu_0 M_S = 0.48$ ), which reflects the presence of exchange interactions between grains. The presence of such interactions is additionally confirmed by S-like shaped behaviour of Wohlfarth plot, which is typically observed in exchange coupled spring magnets [9 - 11].

#### REFERENCES

- [1] J.L. Sanchez Ll, R. Bustamante S., V.M.T.S. Barthem, P.E.V. de Miranda, *Journal of Magnetism and Magnetic Materials* **294**, e131–e135 (2005).
- [2] E.H. Feutrill, Ph. D. Dissertation, The University of Western Australia, 1999.
- [3] E.P. Wohlfarth, *J. Appl. Phys.* **29**, 595 (1958).
- [4] R.A. McCurrie, P. Grunt P, *Philos. Mag.* **13**, 567 (1966).
- [5] E.C. Stoner, E. P. Wohlfarth, *Phil. Trans. Roy. Soc. London* **240**, 599 (1948).
- [6] D. Plusa, M. Dospial, U. Kotlarczyk, B. Slusarek, T. Mydlarz, *Archives of Metallurgy and Materials* **53**, 867-874 (2008).
- [7] M. Dospial, D. Plusa, B. Slusarek; *J Magn Magn Mater* **324** (2012) 843–848
- [8] J. Olszewski, J. Zbroszczyk, M. Hasiak, J. Kaleta, M. Nabialek, P. Brągiel, K. Sobczyk, W. Cieurzyńska, J. Świerczek, A. Łukiewska, *Journal of Rare Earths* **27**, 680-683 (2009).
- [9] A. E. Ceglarek, D. Plusa, M. J. Dośpiał, M.G. Nabialek, P. Pietrusiewicz, *Acta Physica Polonica A*, **121**,5-6, 1278- 1280 (2012).
- [10] D. Plusa, M. Dospial, B. Slusarek, U. Kotlarczyk, T. Mydlarz, *Rev Adv. Mater. Sci.* **18**, 541-544 (2008).
- [11] M. Dospial, D. Plusa, *Journal of Magnetism and Magnetic Materials* **330**, 152–158 (2013).

*Received: 20 September 2014.*