Investigation of the Magnetization Reversal Process of High-Remanent Nd$_{10}$Fe$_{83}$Zr$_{1}$B$_{6}$ Alloy in the As-Cast State

A.E. Ceglarek*, D. Płuża, M.J. Dośpial, M.G. Nabialek and P. Pietrusiewicz

Institute of Physics, Częstochowa University of Technology, al. Armii Krajowej 19, 42-200 Częstochowa, Poland

In this work the magnetic properties of ribbons with composition of Nd$_{10}$Fe$_{83}$Zr$_{1}$B$_{6}$ obtained by using the melt-spinning method were studied. From the X-ray diffraction patterns the phase composition was determined. It was found that investigated alloy was composed of α-Fe and Nd$_{2}$Fe$_{14}$B phases. From the peaks broadening the grain sizes of α-Fe and Nd$_{2}$Fe$_{14}$B phases were estimated as equal to 20 nm and 40 nm, respectively. From the recoil curves the reversible $\mu_0M_{rev}$ and irreversible $\mu_0M_{irr}$ parts of magnetization and differential susceptibility $\chi_{m}$ were determined as a function of an applied field. From these dependences it was found that the pinning of domain walls at the grain boundaries is the main magnetization reversal process. The interactions between grains were investigated by means of the δM plot. It was stated that short range exchange interaction between grains of hard and soft phases are dominant and causes the remanence enhancement.

PACS: 75.30.Et, 75.50.Vw, 75.50.Ww, 75.60.Ej, 75.60.Jk

1. Introduction

The two-phase nanocomposite magnetic materials consisting of hard Nd$_2$Fe$_{14}$B and soft α-Fe or Fe$_2$B nanoscaled grains have been intensively investigated since the first announcements by Coecheckn et al. [1] and Manaf et al. [2] on the remanence enhancement arising from the exchange interaction between the grains with different magnetic hardness. In such alloys the high anisotropy field of hard Nd$_2$Fe$_{14}$B phase is combined with the high saturation magnetization of soft α-Fe or Fe$_2$B phases. Such nanocomposites are very attractive material for production of permanent magnets (called the exchange spring magnets [3]) due to the possibility to obtain the high remanence and consequently the high maximum energy product $(BH)_{max}$. Unfortunately, the presence of soft magnetic phase results in decrease in coercivity. Therefore the high values of maximum energy product predicted by theory are not achieved up till now in spite of many attempts which have been made to improve the microstructure and thus coercivity.

Exchange coupling between neighbouring grains causing the remanence enhancement above $\mu_0M_R = 0.5\mu_0M_S$ reduces simultaneously the coercivity. It is essential that this reduction should not be smaller than $\mu_0H_C = 0.5\mu_0M_R$ in order to get the appropriately high maximum energy product [3, 4].

The melt spinning method is a simple and economical way to prepare the nanocomposite ribbons from alloys with lower amount of expensive Nd in comparison with the stoichiometric one. The nanocomposites can be obtained either by formation of amorphous alloy by rapid quenching of liquid alloy on rotating copper wheel and the crystallization by annealing or directly solidifying the melt. The better magnetic properties can be obtained by the latter method because the possibility of grain growth during annealing in a former one [5].

The large number of experimental investigation on Nd–Fe–Zr–B type compounds have been performed up to now. These articles usually describe the manufacturing conditions and their influence on structure, magnetic properties and thermal stability [6–8]. But none of them were devoted to magnetization reversal processes in Nd$_{10}$Fe$_{83}$Zr$_{1}$B$_{6}$ ribbons.

The aim of this paper was to obtain the exchange coupled nanocomposite ribbons having the good magnetic properties in an as-cast state and discuss the coercivity mechanism in that sample.

2. Material and experimental methods

In this work, the nanocomposites were obtained on the base of Nd$_2$Fe$_{14}$B alloy. The ingots of the alloy were obtained by arc-melting method in the protective argon atmosphere using high purity constituents. Boron was added in the form of an alloy with known composition of Fe-45.4 at.%, B-54.6 at.%. Due to the high ability to oxidize the rare earth, titanium was used as an oxygen absorbent. Small amount of Zr was added in order to hinder the grain growth [6].

The nanocomposite ribbons were prepared by rapid quenching of the liquid alloy on a rotating copper wheel with the linear velocity of 20 m/s and pressure inside of chamber of 0.4 × 10$^5$ Pa. The phase composition was determined using Bruker D8 Advance X-ray diffractometer equipped with Lynx Eye semiconductor counter and Cu $K_\alpha$ a radiation source. The average grain sizes were determined using the Sherrer formula.

The hysteresis and recoil loops measurements were performed by the LakeShore vibration sample magnetometer working at magnetic field up to 2 T in room temperature. From the major hysteresis loop measurements the magnetic parameters i.e.: coercivity $\mu_0H_C$, remanence $\mu_0M_R$, saturation magnetization $\mu_0M_S$, and maximum energy product $(BH)_{max}$ were determined. From the recoil curves measured in both magnetization and demagnetization directions the reversible $\mu_0M_{rev}$ and irreversible $\mu_0M_{irr}$ magnetization and differential suscepti
bility $\chi_{\text{rev}}$ and $\chi_{\text{irr}}$ components were determined. The $\mu_0M'_{\text{irr}}$ is the value of the magnetization when the magnetizing field is reduced to zero along the recoil curve. $\mu_0M'_{\text{rev}}$ is the magnetization when the reversed field applied to previously saturated sample is reduced to zero. The $\mu_0M'_{\text{irr}}(d)$ is defined as \[10\]:
$$\mu_0M'_{\text{irr}}(d) = 0.5\mu_0M_R \text{ needed for obtaining the high maximum energy product.}$$

As it is seen in Fig. 1 the recoil curves for the investigated ribbon are not completely reversible and enclose some areas but they are much smaller than that published for similar samples [13] suggesting the existence of stronger intergrain coupling.

$$\mu_0M'_{\text{irr}}(d) = 0.5\mu_0M_R \text{ needed for obtaining the high maximum energy product.}$$

3. Results and discussion

From the qualitative analysis of X-ray pattern it was found that the investigated alloy is composed of two phases which includes magnetically hard Nd$_2$Fe$_{14}$B and $\alpha$-Fe soft phases. Using the Sherer formula the average grain sizes of both phases were estimated as equal to $(27 \pm 3)$ nm, $(9 \pm 1)$ nm, respectively.

Field dependence of the total magnetization, reversible and irreversible components of total magnetization during (a) the initial magnetization, (b) the demagnetization.

Field dependence of reversible and irreversible magnetization components for both polarization directions of magnetization is shown in Fig. 2a,b. For comparison the reversible and irreversible magnetization in the second quadrant of hysteresis loop were calculated as

$$\mu_0M_{\text{rev}} = -\mu_0M'_{\text{rev}},$$
$$\mu_0M_{\text{irr}} = \frac{(\mu_0M'_{\text{irr}} - \mu_0M'_{\text{irr}})}{2}.$$

The dependences of reversible magnetization component as a function of an applied field in both polarization directions are similar. Its changes are associated with a reversible rotation of magnetization vector in magnetically soft iron grains and magnetically hard Nd$_2$Fe$_{14}$B grains. Small peaks are observed at an applied magnetic field lower than the coercivity of the sample. Its presence may be associated with $\alpha$-Fe and Nd$_2$Fe$_{14}$B grains decoupling.

The dependences of irreversible magnetization component are characteristic for materials in which the main demagnetization mechanism is the pinning of domain walls at the grain boundaries [14]. At an applied magnetizing (or demagnetizing) field lower than the sample coercivity the irreversible magnetization components have the small values (or close to zero). At higher fields the irreversible components of magnetization rise rapidly and exceed the reversible ones.

The changes in magnetization are better seen in a field dependence of differential susceptibilities for both field directions presented in Fig. 3a,b.
At a field lower than the sample coercivity ($< 0.9$ T) the $\delta M(H)$ value is positive and at higher fields changes its sign [16]. Such shape of the $\delta M(H)$ plot is characteristic for materials in which the short-range exchange interaction dominates over the magnetostatic one and supports the magnetization process.

4. Concluding remarks

The nanocomposite Nd$_{16}$Fe$_{80}$Zr$_{1}$B$_{6}$ ribbon is composed of strongly exchange coupled $\alpha$-Fe and Nd$_{2}$Fe$_{14}$B grains. Manufactured ribbons in as-cast state have better magnetic properties (i.e. $(BH)_{\text{max}}$ = 160 kJ/m$^3$ and $\mu_0M_R = 1.05$ T) in comparison to annealed ribbons with similar composition founded in the literature (i.e. $(BH)_{\text{max}}$ = 140 kJ/m$^3$ and $\mu_0M_R = 0.95$ T [7]). The coercivity equal to 0.87 T fulfills the condition $\mu_0H_C > 0.5\mu_0M_R$. The existing of exchange interaction confirmed by positive $\delta M$ is the source of the remanence and $(BH)_{\text{max}}$ enhancement. High value of coercivity results from the small grain sizes (27 nm and 9 nm for $\alpha$-Fe and Nd$_{2}$Fe$_{14}$B phases, respectively). The main mechanism of the magnetization reversal is the pinning of domain walls.

References