Proceedings of XIX International Scientific Conference "New Technologies and Achievements in Metallurgy, Material Engineering, Production Engineering and Physics", Częstochowa, Poland, June 7–8, 2018

# Magnetic Properties and Magnetization Reversal Processes in Nanocrystalline (Pr, Dy)-(Fe, Co)-B Doped Ni and Mn Ribbons

A. Przybył\*

Institute of Physics, Faculty of Production Engineering and Materials Technology, Częstochowa University of Technology, al. Armii Krajowej 19, 42-200 Częstochowa

The magnetization reversal processes responsible for magnetic properties of rapidly solidified ribbons produced from  $Pr_8Dy_1Fe_{60}Co_7Mn_{6-x}Ni_xB_{14}Zr_1Ti_3$  alloys (where x = 0, 3, 6) were investigated. The ingot samples were obtained by arc-melting of high purity constituent elements. The single roll spinning technique was used for producing rapidly solidified ribbons. Subsequently, the ribbons were subjected to annealing at 963 K temperature for 5 min in order to induce the crystallization. The magnetic parameters were determined from magnetic hysteresis loops measured in the external magnetic field up to 2 T at room temperature. The analysis of the magnetization reversal processes in the tested materials was carried out based on measurements of recoil curves and minor hysteresis loops. The series of recoil curves were obtained for the initially saturated and for the thermally demagnetized specimens. The minor hysteresis loops were used to obtain the field dependences of remanence  $J_r$  and coercivity  $_JH_c$ . In order to characterize interactions between grains of crystalline phases the  $\delta M$  plots were also constructed from recoil curves.

DOI: 10.12693/APhysPolA.135.288

PACS/topics: 75.20.En, 91.60.Pn

#### 1. Introduction

A new group of Fe-based bulk glassy alloy widely investigated in recently are Fe–Co–RE–B alloys containing RE elements such as Nd, Dy, Nb, and Pr [1–4]. The good magnetic properties in this group of alloys appear from the exchange interaction between magnetically hard and soft phases. The hard magnetic phase (Pr,Co)<sub>2</sub>Fe<sub>14</sub>B with high anisotropy leads to high coercivity while the soft magnetic phase  $\alpha$ -Fe causes increase of magnetization. The macroscopic magnetic properties of the magnetization reversal process. An experimental method to designate coercivity mechanism and the intergrain interaction is analysis recoil demagnetization curve and  $\delta M(H)$  plot technique.

In an earlier study, the effect of Mn and Ni addition to (Pr-Dy)-(Fe-Co)B-type alloys on the phase constitution and magnetic properties were investigated [5–7].

Therefore the aim of the present work is to investigate the effects of a small Mn and Ni addition on the magnetic properties and magnetization reversal processes in ribbon alloys annealed at 963 K.

#### 2. Samples preparation and experimental methods

The base material with nominal compositions of the  $Pr_8Dy_1Fe_{60}Co_7Mn_{6-x}Ni_xB_{14}Zr_1Ti_3$  (where x = 0, 3, 6)

were made by arc-melting method in protective argon atmosphere from high purity elements and pre-alloyed Fe–B of known composition. In order to get better homogeneity material the samples were re-melted several times. Subsequently, ribbons were prepare by single roll meltspinning technique under the Ar atmosphere at linear velocity of the copper roll surface of 30 m/s. The ribbons were annealed in quartz tubes under protective argon atmosphere at 963 K for 5 min.

In order to determine magnetization reversal processes the rates of irreversible magnetization changes upon the change of external magnetic field H were studied. The series of recoil curves were obtained for the initially saturated samples and for the thermally demagnetized specimens. Moreover, minor hysteresis loops were used to obtain the field dependences of remanence  $J_r$  and coercivity  $_JH_c$ . In order to characterize interactions between grains of crystalline phases the  $\delta M$  plots were also constructed from recoil curves. The room temperature major and minor hysteresis loops, as well as sets of recoil curves were performed using a vibrating sample magnetometer (LakeShore 7307 VSM) in the external magnetic field up to 1600 kA/m.

### 3. Results and discussion

The basic magnetic parameters were determined from the hysteresis loops measured at room temperature in external magnetic fields up to 1600 kA/m. The loops measured for all investigated samples annealed at 963 K of the  $Pr_8Dy_1Fe_{60}Co_7Mn_{6-x}Ni_xB_{14}Zr_1Ti_3$  (where x = 0, 3, 6) alloy are shown in Fig. 1. With an increase of

<sup>\*</sup>e-mail: przybyl@wip.pcz.pl



Fig. 1. The magnetic hysteresis loops measured for  $Pr_8Dy_1Fe_{60}Co_7Mn_{6-x}Ni_xB_{14}Zr_1Ti_3$  (where x = 0 (a), 3 (b), 6 (c)) ribbons. The inset shows the coercivity with the variation of x from 0 to 6.

FABLE I
---------

Magnetic parameters of  $Pr_8Dy_1Fe_{60}Co_7Mn_{6-x}Ni_xB_{14}Zr_1Ti_3$ (where x = 0, 3, 6) ribbons.

x	$_{J}H_{C}$ [kA/m]	$J_r$ [T]	$J_r/J_s$ [-]	$(BH)_{ m max} \ [{ m kJ/m^3}]$
0	780	0.48	0.68	25.5
3	738	0.68	0.72	73.7
6	517	0.72	0.69	50.6

Ni atomic percent the saturation magnetization increase, which could partially result from the more amount of soft magnetic phase. Changes of coercivity with percent content of Ni are shown in the inset of Fig. 1. The magnetic properties, i.e., coercivity  $_{J}H_{C}$ , remanence  $J_{r}$ , energy product  $(BH)_{\text{max}}$  and  $J_r/J_s$  are listed in Table I. For samples containing Mn two-stage demagnetization curve with the characteristic decrease of the magnetization was observed. This shape of the hysteresis loops is typical for two-phase alloys containing both a hard and soft magnetic phases. The maximum values of the coercivity  $_JH_c = 780$  kA/m was obtained for a sample doped with 6 at.% of Mn. However, this sample is characterized by the lowest value of polarization remanence  $J_r = 0.48$  T (Table I). Replacement of part of the Mn atoms by Ni atoms leads to an increase in the value of remanence, but unfortunately the value of coercivity significantly decreases and for 6 at.% Ni amounts to  $_JH_C = 516$  kA/m. The maximum value of maximum energy product  $(BH)_{\text{max}} = 73.7 \text{ kJ/m}^3$  for 3 at.% of Ni was observed.

To understand magnetic reversal processes the magnetic behavior in above samples was investigated. The recoil curves are shown in Fig. 2. These study were used to determine the  $M_{rev}$  dependence curves in the  $M_{irr}$ function, presented in Fig. 3. For all investigated samples



Fig. 2. Magnetization recoil curves measured for the initially saturated nanocrystalline  $Pr_8Dy_1Fe_{60}Co_7Mn_{6-x}Ni_xB_{14}Zr_1Ti_3$  (where x = 0, 3, 6) ribbons.



Fig. 3. Reversible magnetization  $M_{rev}$  as a function irreversible magnetization  $M_{irr}$  for  $Pr_8Dy_1Fe_{60}Co_7Mn_{6-x}Ni_xB_{14}Zr_1Ti_3$  (where x = 0 (a), 3 (b), 6 (c)) ribbons.

the curves  $M_{rev}$  vs.  $M_{irr}$  evidence a shallow minimum value. Such a shape of curves indicates that pinning field  $H_P$  and the nucleation field  $H_N$  are very close to each other. This shows a more complicated magnetic behavior. In these materials the magnetic reversal process is determined by both nucleation new domain and pinning effect on domain wall motion. Some differences between the manner of Ni-doped ribbons do not indicate significant differences in the magnetization reversal processes.

For more accurate analysis the magnetic reversal processes, the minor hysteresis loops for ribbons initially in the demagnetized state were measured. The normalized dependences of the coercivity  ${}_{J}H_{c}/{}_{J}H_{C}^{\max}$  and remanence  $J_{r}/J_{r}^{\max}$  in the maximum applied magnetic field H curves are shown in Fig. 4. In low fields both coercivity and retentivity for all investigated samples remain at a constant low level. At an applied magnetic field about 500 kA/m in samples containing Ni rapid increase measured values is observed. In the case of sample  $Pr_8Dy_1Fe_{60}Co_7Mn_6B_{14}Zr_1Ti_3$  for the rapid increase of both coercive and remanence a higher value of the external magnetic field is required.



Fig. 4. Initial magnetization curve and normalized field dependences of coercivity  $_JH_C$  and remanence  $J_r$  for  $Pr_8Dy_1Fe_{60}Co_7Mn_{6-x}Ni_xB_{14}Zr_1Ti_3$  (where x = 0 (a), 3 (b), 6 (c)) ribbons.

Normalized coercivity increases a bit faster than remanence, however, these differences are insignificant, which confirms the complex nature of the coercivity mechanism with a slight dominant domain pinning over the nucleation mechanism. This dependence can be seen in samples containing only Mn or only Ni. In the alloy containing both of these elements, the increase in coercivity and remanence is similar.

The magnetization reversal processes are close dependent upon the intergranular exchange coupling. In order to confirm the existence of exchange coupling between grains in the studied materials, the Henkel plots analysis was used. The expression of  $\delta M$  is defined as  $\delta M = m_d(H) - [1 - 2m_r(H)]$  (where  $m_d$  is the reduced demagnetization remanence and  $m_r$  is the reduced magnetization remanence) [8]. The  $\delta M$  curve with a positive peak indicates that exchange coupling is predominant over the magnetostatic interaction, which is associated with the negative deviation.

The  $\delta M(H)$  plots for all tested samples are shown in Fig. 5. A positive peak occurring around the coercive field was observed for all samples. For the alloy containing both Mn and Ni, the peak height is twice as high as for the other two. It is this sample that has the highest value  $(BH)_{\rm max}$ . The low value of  $\delta M$  in samples containing only Mn or only Ni indicates a weak intergranular exchange coupling in these materials. It is very interesting that the introduction of 3% at Ni into the alloy causes a significant increase in the maximum value of  $\delta M$ , which indicates the increase of the importance of intergranular exchange coupling over magnetostatic, resulting in a higher value of  $(BH)_{\rm max}$ .



Fig. 5.  $\delta M(H)$  plots of the Pr<sub>8</sub>Dy<sub>1</sub>Fe<sub>60</sub>Co<sub>7</sub>Mn<sub>6-x</sub>Ni<sub>x</sub>B<sub>14</sub>Zr<sub>1</sub>Ti<sub>3</sub> (where x = 0, 3, 6) ribbons.

#### 4. Conclusions

In this study, the magnetization reversal proresponsible for magnetic properties cesses of  $Pr_8Dy_1Fe_{60}Co_7Mn_{6-x}Ni_xB_{14}Zr_1Ti_3$  (where x =0. 3, 6) ribbons were investigated. The magnetic material was prepared by the melt-spinning technique and then subjected to a short-term heat treatment at 963 K. It has been found that the largest coercivity has an alloy containing 6 at.% Mn, while the highest value of  $(BH)_{max}$  is characterized by a sample containing both Mn and Ni. For all the tested alloys the enchancement of remanence  $(J_r/J_s > 0.5)$  were observed, which indicates the occurrence exchange-coupling in these materials.

In addition, it was found that the magnetization reversal process in these materials has a complex character and is determined by both nucleation new domain and pinning effect on domain wall motion.

## References

- W. Zhang, M. Matsusita, C. Li, K. Kimura, A. Inoue, *Mater. Trans. JIM* 42, 2059 (2001).
- [2] I. Betancourt, H.A. Davies, *Mater. Sci. Technol.* 26, 5 (2010).
- [3] P. Pawlik, K. Pawlik, H.A. Davies, W. Kaszuwara, J.J. Wysłocki, *J. Magn. Magn. Mater.* **316**, e124 (2007).
- [4] R.K. Murakamia, H.R. Rechenberg, A.C. Neiva, F.P. Missell, V. Villas-Boas, J. Magn. Magn. Mater. 320, e65 (2008).
- [5] A. Przybył, K. Pawlik, P. Pawlik, P. Gębara, J.J. Wysłocki, *J. Alloys Comp.* **536S**, S333 (2012).
- [6] A. Przybył, K. Pawlik, P. Pawlik, P. Gębara, J.J. Wysłocki, *Acta Phys. Pol. A* **127**, 579 (2015).
- [7] A. Przybył, K. Pawlik, P. Pawlik, P. Gębara, W. Kaszuwara, J.J. Wysłocki, K. Filipecka, *Acta Phys. Pol. A* **128**, 94 (2015).
- [8] P.E. Kelly, K. O'Grady, P.I. Mayo, R.W. Chantrell, *IEEE Trans. Magn.* 25, 3881 (1989).