Vol. **131** (2017) *ACTA PHYSICA POLONICA A* No. 5

12th Symposium of Magnetic Measurements and Modeling SMMM'2016, Częstochowa–Siewierz, Poland, October 17–19, 2016

Thermomagnetic Properties and First-Order Reversal Curve Analysis of Annealed Fe–Co–Si–B–Mo–P Alloy

M. Hasiak[a,*] and M. Miglierini[b,c]

[a]Department of Mechanics, Materials Science and Engineering, Wrocław University of Science and Technology,
M. Smoluchowskiego 25, 50-370 Wrocław, Poland

[b]Slovak University of Technology in Bratislava, Faculty of Electrical Engineering and Information Technology,
Institute of Nuclear and Physical Engineering, Ilkovičova 3, 812 19 Bratislava, Slovakia

[c]Department of Nuclear Reactors, Faculty of Nuclear Sciences and Physical Engineering,
Czech Technical University in Prague, V Holešovičkách 2, 180 00 Prague, Czech Republic

The paper presents thermomagnetic features and characterization of magnetic interactions in the $Fe_{51}Co_{12}Si_{16}B_8Mo_5P_8$ metallic glass after annealing at 798 K for 1 h. The first-order reversal curve analysis was used to investigate hysteresis curves which provide a more precise estimation of the strength of interactions. The presence of magnetically distinct regions was revealed.

DOI: 10.12693/APhysPolA.131.1222

PACS/topics: 75.50.Bb, 75.60.–d, 75.50.Tt

1. Introduction

Recently, amorphous Fe–Si–B–P–Cu soft magnetic material (NANOMET) [1, 2] as well as alloys containing Co atoms [3] are intensively studied. They exhibit interesting soft magnetic characteristics, i.e. high magnetic saturation, high magnetic permeability, and extremely low coercivity which are required for magnetic cores. One of the most important parameters in the formation of a nanocrystalline structure in Fe–Si–B–P–Cu alloys is the heating rate [4]. Heat treatment with high heating leads to the formation of an ultrafine structure whereas a low heating rate produces non-uniform structure and deterioration of magnetic properties. The aim of this work is to study magnetic properties and magnetic interactions in the $Fe_{51}Co_{12}Si_{16}B_8Mo_5P_8$ metallic glass after annealing in a vicinity of the crystallization temperature by using the first-order reversal curve (FORC) method [5].

2. Experimental details

Amorphous $Fe_{51}Co_{12}Si_{16}B_8Mo_5P_8$ (at.%) alloy was prepared using a rapid solidification method on a copper quenching wheel. The resulting ribbon was about 0.02 mm thick and 6 mm wide. Its nominal chemical composition was confirmed by inductively coupled plasma mass spectroscopy (ICP-MS) and atomic absorption spectroscopy (AAS). The crystallization temperature was determined from a DCS curve recorded with a heating rate of 10 K/min in a temperature range from room temperature (RT) to 1173 K. Annealing of the as-quenched alloy was performed at 798 K for 1 h in argon atmosphere. The heating rate in annealing process of the sample in the furnace from room temperature to 798 K was also 10 K/min. The amorphicity and degree of crystallinity of the as-quenched and annealed samples was examined by X-ray diffractometry performed at room temperature. Magnetic characteristics were measured with the help of VersaLab system (Quantum Design) in temperature range 50–400 K. Magnetization (M) as a function of external magnetic field (H) and/or temperature was recorded below the Curie point. Hysteresis loops as well as first-order reversal curves were measured as a set of $M(H)$ curves. Evaluation of the obtained results was performed by the FORCinell software [6]. All measurements were performed upon the annealed sample.

3. Results and discussion

Microstructure and crystallization kinetics studies of the $Fe_{51}Co_{12}Si_{16}B_8Mo_5P_8$ alloy are described in more detail in our previous papers [7–9]. The analysis of DSC characteristic recorded in the temperature range 300–1173 K for the as-quenched $Fe_{51}Co_{12}Si_{16}B_8Mo_5P_8$ precursor shows two distinguished exothermic peaks which indicate presence of a two-step crystallization [7]. The primary and secondary crystallization temperatures were determined as 839 K and 963 K, respectively. Moreover, an additional small kink at about 922 K can be seen.

It is worth noting that microstructure analysis performed by X-ray diffraction measurements with Co K_α radiation of the as-quenched $Fe_{51}Co_{12}Si_{16}B_8Mo_5P_8$ sample shows only a wide halo which is characteristic for amorphous materials. The maximum of a broad diffraction peak is positioned at the 2Θ angle of about 55 deg [7]. The X-ray diffraction pattern taken from the annealed sample shows the presence of several narrow peaks with low intensity. They can be assigned to Fe-free crystalline phases [7] which start to appear even though the first crystallization temperature, as derived from DSC, is still not reached. As a consequence, modification of the magnetic microstructure can also be expected.

---

*corresponding author; e-mail: Mariusz.Hasiak@pwr.edu.pl

Thermomagnetic characteristics obtained for the annealed $Fe_{51}Co_{12}Si_{16}B_8Mo_5P_8$ alloy in zero-field cooled (ZFC) and field cooled (FC) regimes are presented in Fig. 1. Temperature dependences of magnetization measured in ZFC mode in Fig. 1 (upper figure) show that for an external magnetic field up to 250 mT the magnetization slightly increases with temperature up to its maximum and then suddenly drops down in the vicinity of Curie temperature. Moreover, with an increasing external magnetic field, a growth of magnetization and a shift of its maximum (blocking temperature T_B) towards lower temperatures is observed. The blocking temperatures are 383, 343, and 244 K for the $M(T)$ curves recorded in ZFC mode at external magnetic field of 50, 100, and 250 mT, respectively.

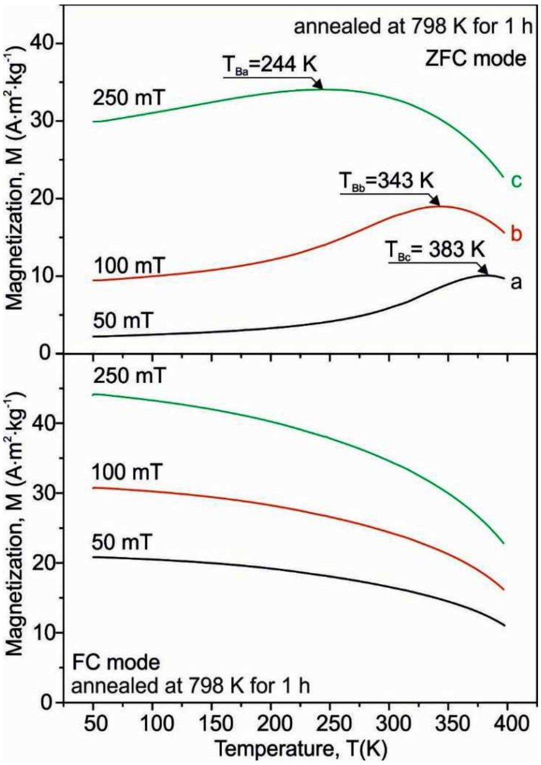

Fig. 1. ZFC (upper figure) and FC (lower figure) magnetization versus temperature for the annealed $Fe_{51}Co_{12}Si_{16}B_8Mo_5P_8$ alloy measured in external magnetic fields of 50, 100, and 250 mT.

Figure 1 (lower figure) shows FC magnetization versus temperature for the annealed $Fe_{51}Co_{12}Si_{16}B_8Mo_5P_8$ alloy measured in the indicated external magnetic fields. It is seen that the magnetization falls monotonically with temperature in all fields. $M(T)$ dependences obtained under ZFC and FC conditions suggest that magnetic properties of the investigated material depend on cooling conditions, and they must be taken into account in the design process of electrical devices.

Representative hysteresis loops recorded for the $Fe_{51}Co_{12}Si_{16}B_8Mo_5P_8$ alloy annealed at 798 K for 1 h in temperature range from 100 to 400 K with a step of 100 K and a maximum external magnetic field of 50 mT are presented in Fig. 2. It is seen that with increasing temperature of measurement, the magnetization decreases as it is also visible from $M(T)$ curves in Fig. 1 (lower figure). The magnetization measured in an external magnetic field of 50 mT for the sample measured at 400 K is equal to half the value of magnetization for material measured at 100 K. Moreover, changes in magnetic structure are represented by a decrease of coercive field with temperature. The obtained coercive fields recorded at 100, 200, 300, and 400 K for the investigated material equal to 8.98, 8.44, 5.21, and 1.16 mT, respectively.

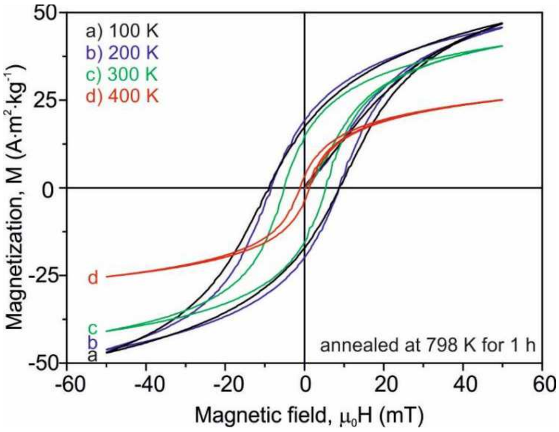

Fig. 2. Hysteresis loops for the annealed $Fe_{51}Co_{12}Si_{16}B_8Mo_5P_8$ alloy measured in external magnetic field of 50 mT and the indicated temperatures.

Magnetic characterization of single-phase magnets, i.e. magnetic interactions in materials with high value of coercive field, is usually performed by investigations of hysteresis loops. A problem appears when complex magnetic interactions are present in a multi-phase material. The first-order reversal curve (FORC) analysis is one of the methods which can provide information about interaction field between such multiphase magnetic particles/phases. FORCs are a specific class of minor hysteresis loops. A more detailed description of the principles of FORC can be found elsewhere [5]. This method was used to study thermally induced decoupling of nanosized crystallites embedded in an amorphous matrix of a Co–Si–B–Fe–(Mo–Ni) [10] and Fe–Zr–B–Cu [11, 12] alloys. Here, we employ FORC analysis for a study of annealed $Fe_{51}Co_{12}Si_{16}B_8Mo_5P_8$ alloy which is just at the onset of crystallization.

The set of the FORCs recorded at 77 K is presented in Fig. 3. They were successively recorded as minor curves. The sample is exposed to an initial positive saturation field B_{sat} (= 0.3 T) which is reduced down to a reversal field B_b. The applied field B_a is then increased up to B_{sat} while measuring the magnetization $M(B_a, B_b)$. This procedure is repeated for a new B_b value that differs by ΔB_b from the previous one. Precision of the method depends upon ΔB_b, i.e. the total number of curves which must cover the whole hysteresis area. In our case, 123 minor curves were recorded.

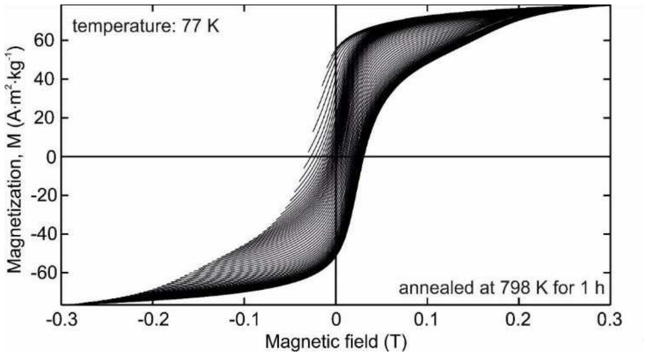

Fig. 3. Set of first-order reversal curves for the $Fe_{51}Co_{12}Si_{16}B_8Mo_5P_8$ alloy after annealing at 798 K for 1 h measured at 77 K in a maximal external magnetic field of 0.3 T.

FORC distribution $\rho(B_a, B_b)$ is defined as the second derivative of the magnetization M [5]:

$$\rho\left(B_a, B_b\right) = -\frac{1}{2}\frac{\partial^2 M\left(B_a, B_b\right)}{\partial B_a \partial B_b}$$

where B_a and B_b are the magnetization field and reversal field, respectively. FORC distribution experimentally characterizes the complete irreversible magnetic behavior of a given system. It is usually represented as a contour plot in a Preisach plane plotted in B_c and B_u ordinates. The latter are calculated according to the transformations: $B_c = (B_a - B_b)/2$ and $B_u = (B_a + B_b)/2$. The coercivity axis B_c and the interactions field axis B_u are directly related to the local irreversible properties. The FORC distribution with the B_c and B_u axes represents the statistical distribution of irreversible processes related to their local coercivity and bias (interaction field) values. Using this technique, one can decouple the irreversible behavior which stems from magnetically different phases in the investigated system.

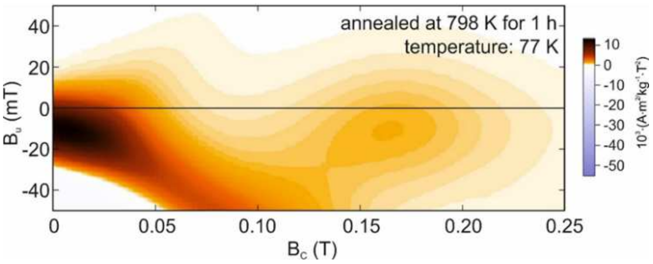

Fig. 4. FORC diagram for the $Fe_{51}Co_{12}Si_{16}B_8Mo_5P_8$ metallic glass after annealing at 798 K for 1 h measured at 77 K in a maximal external magnetic field of 0.3 T.

The shapes of the hysteresis loops in Fig. 3 indicate that they can be decomposed into two loops with different coercive fields. Indeed, complex magnetic behavior with two different distributions can be clearly distinguished in Fig. 4. Here, the FORC diagram for the annealed $Fe_{51}Co_{12}Si_{16}B_8Mo_5P_8$ alloy measured at 77 K is shown. One well pronounced maximum of coercivity is visible at $B_c \approx 0.01$ T whereas another local maximum appears at $B_c \approx 0.17$ T. The former can be associated with softer magnetic phase that corresponds to the residual amorphous matrix. The second region of microcoercivity with higher B_c can be assigned to the presence of a small number of magnetically hard crystallites. The interaction field values are $B_u \approx$–10 mT.

4. Conclusions

Thermomagnetic properties and magnetic interactions were studied in $Fe_{51}Co_{12}Si_{16}B_8Mo_5P_8$ alloy annealed at temperature which is about 40 K smaller than the first crystallization temperature determined from DSC. The presence of a small number of crystallites was revealed by XRD and seems to be connected with low temperature crystallization. They have also significantly affected the resulting magnetic microstructure which exhibited complex behaviour. However, the discrimination between contributions from each magnetically ordered phase to the hysteresis loops is not so straightforward. In order to investigate the origin of the observed magnetic interactions, we have employed the FORC analysis. Two magnetic phases with microcoercivity values of 0.01 T and 0.17 T were revealed. The former softer phase corresponds to the residual amorphous matrix and the latter represents magnetically harder crystallites.

Acknowledgments

Financial support of the grant VEGA 1/0182/16 is acknowledged.

References

[1] M. Chiba, A. Urata, H. Matsumoto, S. Yoshida, A. Makino, *IEEE Trans. Magn.* **47**, 2845 (2011).

[2] A. Makino, *IEEE Trans. Magn.* **48**, 1331 (2012).

[3] K. Takenaka, A.D. Setyawan, Y. Zhang, P. Sharma, N. Nishiyama, A. Makino, *Mater. Trans.* **56**, 372 (2015).

[4] P. Sharma, X. Zhang, Y. Zhang, A. Makino, *Scr. Mater.* **95**, 3 (2015).

[5] Ch.R. Pike, A.P. Roberts, K.L. Verosub, *J. Appl. Phys.* **85**, 6660 (1999).

[6] R.J. Harrison, J.M. Feinberg, *Geochem. Geophys. Geosyst.* **9**, Q05016 (2008).

[7] N. Amini, M. Miglierini, M. Hasiak, *AIP Conf. Proc.* **1781**, 020001 (2016).

[8] M. Cesnek, M. Miglierini, T. Kmječ, J. Kohout, N. Amini, D. Janičkovič, P. Matúš, *Acta Phys. Pol. A* **131**, 744 (2017).

[9] M. Hasiak, M. Miglierini, N. Amini, M. Bujdoš, *Nukleonika*, 2017, in press.

[10] J.C. Martínez-García, M. Rivas, J.A. García, *Physica B* **486**, 84 (2016).

[11] F. Béron, L.A.S. de Oliveira, M. Knobel, K.R. Pirota, *J. Phys. D Appl. Phys.* **46**, 045003 (2013).

[12] M. Rivas, J.C. Martínez-García, P. Gorria, *J. Magn. Magn. Mater.* **400**, 315 (2016).