Influence of Co Doping on Induced Anisotropy and Domain Structure in Magnetic Field Annealed \((\text{Fe}_{1-x}\text{Co}_x)_{79}\text{Mo}_8\text{Cu}_1\text{B}_{12}\)

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The effect of heat treatment in applied magnetic field on the induced anisotropy and domain structure of \((\text{Fe}_{1-x}\text{Co}_x)_{79}\text{Mo}_8\text{Cu}_1\text{B}_{12}\) \((x = 0, 0.2, 0.5)\) nanocrystalline alloy system was investigated. A heat treatment of Co-doped samples under the application of longitudinal magnetic field resulted in squared hysteresis loops characterized by very low coercive field values. Sheared loops with tunable slope and good field linearity were obtained after annealing in transverse magnetic field. Corresponding domain structure showed uniform character, oriented in the direction parallel or perpendicular to the ribbon axis after longitudinal or transverse magnetic field annealing, respectively. No effect of magnetic field annealing was found in Co-free sample. Correlations between Co-doping, the Curie temperature, and soft magnetic properties after magnetic field annealing are discussed.

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1. Introduction

Amorphous and nanocrystalline FeMoCuB alloys show good soft magnetic behaviour; however, the low Curie temperature limits their possible applications at elevated temperatures [1, 2]. Partial substitution of Fe by Co [3] leads to enhancement of the Curie temperature, and consequently improves thermal stability of ferromagnetic state in these alloys.

Heat treatment of ferromagnetic alloys in applied magnetic field is an effective tool for tailoring their magnetic properties [4, 5]. In many soft magnetic materials including various nanocrystalline and amorphous alloys, the phenomenon of field annealing and induced anisotropy is often explained in terms of magnetic atoms pair ordering mechanism. This mechanism is particularly efficient when the sample remains in ferromagnetic state during thermal processing and at the same time the thermal energy of atoms is sufficiently high to allow their ordering [6].

In this paper, impact of the magnetic field annealing on nanocrystalline system \((\text{Fe}_{1-x}\text{Co}_x)_{79}\text{Mo}_8\text{Cu}_1\text{B}_{12}\) \((x = 0, 0.2, 0.5)\) is investigated. Obtained results provide better understanding of the relation between thermal stability of ferromagnetic state and strength of induced anisotropy after magnetic field annealing.

2. Experimental

The amorphous ribbons of \((\text{Fe}_{1-x}\text{Co}_x)_{79}\text{Mo}_8\text{Cu}_1\text{B}_{12}\) \((x = 0, 0.2, 0.5)\) series were produced by planar flow casting. Samples were isothermally annealed under a high vacuum for 1 h at 703 K. In order to create preferred orientation of directional order, annealing process took place in presence of the longitudinal magnetic field (LF) of 40 kA/m, and transverse magnetic field (TF) of 640 kA/m. After treatment, specimens were field cooled to a room temperature. The reference samples were also annealed and cooled under the same thermal conditions in zero magnetic field (ZF). Microstructure of annealed samples was investigated by the transmission electron microscopy (TEM) and X-ray diffraction (XRD). To obtain temperature dependence of magnetization, a vibrating sample magnetometer (VSM) was used for measurements above the room temperature, up to 870 K. SQUID magnetometer was, on the other hand, used to acquire the magnetization values at low temperatures. Soft magnetic behaviour of amorphous and annealed samples was investigated by the Forster type \(B-H\) loop tracer. The magneto-optical Kerr microscopy was used for observation of the magnetic domain structure.

3. Results and discussion

Microstructure investigations have revealed formation of the nanocrystalline bcc-Fe, or bcc-FeCo grains, embedded in the amorphous matrix for annealed Co-free and Co-doped samples, respectively. Acquired TEM micrographs in Fig. 1 show that the crystallization was slightly less developed in \(\text{Fe}_{79}\text{Mo}_8\text{Cu}_1\text{B}_{12}\) samples, compared to Co-doped ones. We note that the microstructure characteristics in studied alloys were not influenced by presence of the external magnetic field, as compared to the ZF-annealed reference samples.

Figure 2 shows temperature dependences of magnetization of the as-quenched samples. It is evident that

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Co-free alloy exhibits ferro-paramagnetic transition below the room temperature. The Curie temperature value $T_C(\text{am})$, determined as the minimum of derivative $\delta M/\delta T$ in temperature range from 200 to 300 K, equals to $\approx 252$ K. Increasing Co content shifts $T_C(\text{am})$ to the higher temperature range. The abrupt increase of magnetization above 600 K is related to the primary crystallization of ferromagnetic nanocrystalline grains with higher Curie temperature.

**Fig. 1.** Microstructure of $(\text{Fe}_{1-x}\text{Co}_x)_{79}\text{Mo}_8\text{Cu}_1\text{B}_{12}$: (a) $x = 0$, (b) $x = 0.2$, (c) $x = 0.5$, after annealing at 703 K for 1 h.

**Fig. 2.** Thermomagnetic plots of the as-quenched samples.

Figure 3 presents hysteresis loops, obtained after different field annealing and measured at the room temperature. As can be seen in Fig. 3a no effects of field annealing are visible in the Co-free alloy. Here, paramagnetic state during heat treatment and presence of sole ferromagnetic element (Fe) did not support preferred atomic pair orientation, essential for creation of the field induced anisotropy [6]. In addition, the ferromagnetic bcc-Fe grains are embedded in relatively weak magnetic amorphous residual phase, which weakens coupling between individual grains, and markedly increases the coercivity value [7].

Improved stability of ferromagnetic state towards high temperatures and presence of two kinds of ferromagnetic elements in composition enabled process of magnetic atoms pair ordering in Co-doped alloys. In both cases LF annealing increased the squareness of the loops with accompanied coercivity reduction, while sheared loops with linearity up to anisotropy field resulted from TF annealing. One can clearly see from comparison of Fig. 3b and c that the effects of field annealing are more pronounced in the equiatomic FeCo alloy, which maintains its ferromagnetic character during the whole process of heat treatment. In the case of $(\text{Fe}_4\text{Co}_1)_{79}\text{Mo}_8\text{Cu}_1\text{B}_{12}$, sample with the Curie temperature lower than annealing temperature, the atomic pair ordering mechanism was operative in limited temperature range, mainly during cooling process below $T_C$, where thermal rearrangement of atomic-pairs to the direction of the applied magnetic field was less efficient. The stronger induced anisotropy in field annealed equiatomic FeCo alloy is accompanied by a marked decrease of their coercive field. The $H_c$ value of 3 A/m obtained after LF annealing is comparable with the lowest $H_c$ values reported for HITPERM alloys so far [8, 9].

**Fig. 3.** Hysteresis loops of $(\text{Fe}_{1-x}\text{Co}_x)_{79}\text{Mo}_8\text{Cu}_1\text{B}_{12}$ ($x = 0, 0.2, 0.5$) before and after different field annealing treatment, performed for 1 h at 703 K.

Figure 4 shows domain patterns of samples with Co content $x = 0.5$ in the as-quenched state (a) and after ZF thermal treatment (b). Annealing without applied magnetic field led to stabilization of complicated domain
structure consisting of wide primary domains and maze-shaped secondary domains, characteristic for the amorphous state. The operative mechanism responsible for this stabilization is closely related to self-magnetic annealing process [4, 8]. On the other hand, as presented in Fig. 5, heat treatment in the magnetic field resulted in formation of uniform domain structure. Here, a simple structure consisting of wide domains oriented parallel to the ribbon axis is obtained after LF treatment. On the other hand, narrower domains aligned vertical to the ribbon axis are product of processing under TF conditions. Better uniformity of domain patterns observed in the equiatomic FeCo sample may be evidence of stronger response of this alloy to magnetic field annealing.

4. Conclusions

In this work, induced anisotropy and domain structure were studied in a system of nanocrystalline alloys (Fe$_{1-x}$Co$_x$)$_79$Mo$_8$Cu$_1$B$_{12}$ with different Co/Fe ratio. We have shown that effectiveness of thermal processing in applied magnetic field is linked to the temperature stability of ferromagnetic state of the soft magnetic material. Heat treatment of formerly paramagnetic Co-free alloy resulted in high coercivity hysteresis behaviour at the room temperature. Nearly identical shapes of loops indicate no observable formation of induced anisotropy despite the presence of the magnetic field during annealing process. Co addition increases the Curie temperature and supports creation of directional order. LF annealing therefore results in squared hysteresis loops and accompanies reduction of coercivity values, whereas sheared hysteresis loops are achieved by TF annealing. Respective domain patterns are less uniform in the samples with lower Co content, compared to the equiatomic FeCo composition, which may demonstrate weaker effect of the atomic pair ordering.

Acknowledgments

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