Improvement of soft magnetic properties in Fe-based rapidly quenched alloys and FeSi electrical steels by magnetic field annealing

Ivan Škorvánek - Jozef Marcin - Jozef Kováč - Peter Švec - František Kováč

In this work, a controllable field-induced magnetic anisotropy is produced in series of Fe-Co-Mo-B type amorphous and nanocrystalline alloys with different amount of crystalline phase. The amorphous melt-spun ribbons were isothermally annealed under a high vacuum at temperatures 713 K ≤ Ta ≤ 823 K in the presence of transverse or longitudinal static magnetic field, which was sufficient to reach their full magnetic saturation. The reference samples were annealed and cooled in a zero magnetic field. The structural analysis has revealed formation of a two-phase nanocrystalline microstructure with ultrafine BCC-FeCo grains embedded in an amorphous matrix. The zero field annealed specimens show an appreciable increase of the coercivity with nanocrystallization. Sheared loops with good field linearity and improved magnetic softness were achieved for all investigated alloys after transverse field annealing. Heat treatment in longitudinal field results in squared hysteresis loops is characterized by extremely low coercive field values in the range of 3-8 A/m. These values are superior for HITPERM-type alloys and they remain fairly stable also at elevated temperatures. The magnetic field annealing effects were investigated also in the series of non-oriented electrical steel sheets containing about 2.9 wt.% Si characterized by a different degree of deformation. Here, the observed improvement of soft magnetic properties due to heat treatment in static magnetic field is less significant mainly because of the higher magnetocrystalline anisotropy. The development of induced anisotropy in the field-annealed alloys is discussed in the frame of the magnetic atoms pair ordering theory.

K e y w o r d s: soft magnetic properties, magnetic annealing, rapidly quenched alloys, FeSi electrical steels

1 INTRODUCTION

The reduction of the grain sizes to the nanometer range may drastically vary the functional properties of materials, including the magnetic behavior. Typical examples of such systems are the nanocrystalline, Fe-based alloys prepared by devitrification of melt-spun amorphous precursors, which belong to an important group of soft magnetic materials [1]. The properties of these materials can vary widely, depending on the size and volume fraction of the nanocrystalline grains as well as on the magnetic properties of the intergranular amorphous matrix. It has been shown that the crucial role in the improvement of their soft magnetic behaviour is played by the reduction of the effective magnetic anisotropy, which occurs when the size of nanocrystals become comparable with the magnetic exchange length [2].

The discovery of the excellent soft magnetic properties in the nanocrystalline alloys based on FeCuNbSiB, and Fe(Zr,Nb)B(Cu), called FINEMET and NANOPERM, as well as the later development of the nanocrystalline (Fe,Co)MBCu alloys (M=Zr, Nb and Hf), named HITPERM, has stimulated an enormous research activity in these systems [1]. The HITPERM alloys, display usually less favorable soft magnetic properties as compared to the FINEMET or NANOPERM alloys. However, they exhibit a higher saturation magnetic flux density and they are capable of operation at higher temperatures [3].

In order to enhance the application potential of the FeCo-based nanocrystalline alloys it is important to deepen knowledge about the available processing techniques that can be used to tailor their magnetic properties. One possible way, which could be employed for this purpose, is the thermal processing under the presence of external magnetic field, called also „magnetic annealing“. The effect of heat treatment under the presence of magnetic field is to superpose on the material an extra annealing-induced magnetic anisotropy in addition to whatever anisotropies that may have been present originally. This induced anisotropy is almost always uniaxial, i.e. it creates an easy axis of magnetization, which complies with the direction of magnetization during annealing. In case of soft magnetic nanocrystalline alloys, it was shown that the local random magnetocrystalline anisotropies are strongly suppressed by exchange interactions [4], and thus, they can be easily overcome by the long ranged macroscopic anisotropy induced by field annealing.

Our previous study on the magnetic field annealing effects in the FeCoNbB-type nanocrystalline alloys with various ratios of Fe/Co atoms has clearly demonstrated that the improvement of the soft magnetic characteristics due to field annealing is most significant for the Fe1-xCox concentrations close to x=0.5 [5,6]. Such behavior strongly indicates that the operative mechanism of induced anisotropy in these alloys is the magnetic atoms pair ordering.

In this work, a controllable field-induced magnetic anisotropy is produced in the series of nanocrystalline $Fe_{38}Co_{38}Mo_8B_{15}Cu$ samples with different amount of crystalline phase. We report on the beneficial effects of both longitudinal and transverse magnetic field applied during the heat treatment process on the application-oriented magnetic characteristics of these soft ferromagnets.

2 EXPERIMENTAL PART

Master alloys have been prepared by arc-melting from elements of 99.95 % purity. Amorphous ribbons 6 mm wide and ~ 25 μm thick were produced by planar flow casting. Chemical composition of the ribbons was checked by inductionally coupled plasma spectrometer and found to be as indicated to the accuracy of 3 % of the nominal content of each element. In order to prepare the nanocrystalline samples with preferred direction of induced anisotropy, the pieces of amorphous ribbons (6 cm long) were isothermally annealed under a high vacuum for 1 hour at different temperatures above the crystallization temperature in the presence of transverse (TFA) or longitudinal (LFA) magnetic field. In the case of TFA-annealed samples (Fig. 1(a)), the furnace was placed inside the commercial permanent magnet system (Magnetic Solutions LTD) producing a magnetic field of 640 kA/m directed in the plane of the ribbon and perpendicular to its length. In case of LFA-annealed samples (Fig. 1(b)), the furnace was inserted into the water-cooled solenoidal coil that provided a magnetic field of 20 kA/m oriented along the ribbon length. After such annealing, the specimens were slowly cooled to room temperature in a presence of the magnetic field. A typical cooling rate was 3 K/min. The reference samples were annealed and cooled in a zero magnetic field (ZFA) under the same conditions.

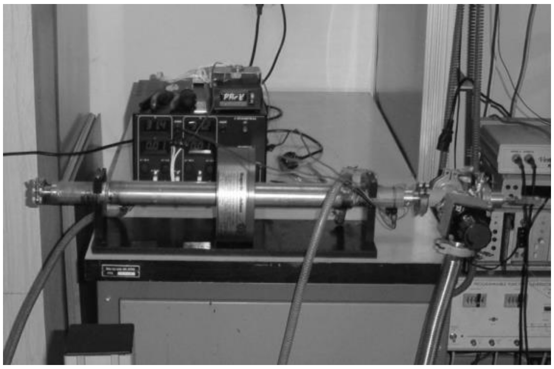

Figure 1 (a). Setup for transverse field annealing.

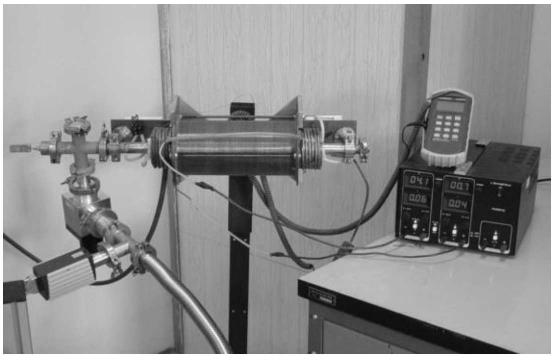

Figure 1 (b). Setup for longitudinal field annealing.

Changes of microstructure upon annealing were investigated by transmission electron microscopy (TEM). Samples for transmission electron microscopy were thinned, after corresponding heat treatment, by ion beam milling, TEM and electron diffraction observations were performed using JEM1200 EX microscope. The soft magnetic behavior was investigated by using a Forster type B-H loop tracer.

3 RESULTS AND DISCUSSION

DSC thermograms for the as-quenched samples have revealed the presence of the characteristic exothermal peaks with onset at T_{x1} = 695 K and the peak position at T_{p1} = 719 K that correspond to the formation of the nanocrystalline bcc-FeCo. The changes in microstructure upon annealing were examined by TEM. Ultrafine grains were observed after partial crystallization of $Fe_{38}Co_{38}Mo_8B_{15}Cu$ with typical grain dimensions ranged from 4-8 nm. No influence of an applied field on grain size or texture was found.

A main attention of this work has been devoted to the study of the effects of annealing under presence of external magnetic field in order to induce controllable uniaxial anisotropy in the samples. The effect of field annealing on the hysteresis loops of $Fe_{38}Co_{38}Mo_8B_{15}Cu$ alloys is demonstrated in Fig. 2. The shape of the hysteresis loop is dictated by the relative importance of domain wall displacement and magnetic moment rotation processes in the sample. According to direction of the induced anisotropy, the magnetization curves with large or small squareness ratio could be obtained. The rotation processes tend to dominate after annealing in the transverse magnetic field, and consequently, the sheared loops with relatively good field linearity are achieved. Such characteristics are of particular interest for high frequency transformers and magnetic sensors. A heat treatment under the presence of longitudinal magnetic field results in squared hysteresis loops that are characterized by a significant reduction of the coercivity. The coercive field values for LF annealed samples are in the range of 3 – 8 A/m, i.e. they are markedly lower than those previously reported for the field annealed HITPERM-type alloys [6-9].

From the area in the first quadrant between the loops corresponding to TFA samples (hard direction) and LFA samples (easy direction), the values of induced anisotropy constant, K_u, can be determined. The maximum value of induced anisotropy constant $K_u \sim 935$ J/m^3 is observed for the $Fe_{38}Co_{38}Mo_8B_{15}Cu$ annealed at 743 K. Directional order theory predicts the dependence of induced anisotropy for binary alloys with two constituent magnetic elements A_xB_{1-x} to go as $x^2(1-x)^2$ [9]. The composition of the nanocrystalline alloys studied in the present paper are very close to the equiatomic FeCo concentration, which explains the observed strong influence of the magnetic field annealing treatment. Directional ordering effects can occur even if the alloy is heat treated below its Curie temperature in the absence of an external magnetic field. In this case, the internal magnetic field of each domain will influence the directionality of diffusion. The consequence of this „self magnetic annealing“ is in that the domains and domain walls tend to be stabilized in the positions they occupied during the annealing, which results often in undesirable increase of coercive field. The fact that the field annealed samples reveal a smaller coercivity than the samples annealed without field can thus be understood from more simple domain configuration due to the uniform induced anisotropy, which in addition suppress the effects of the angular dispersion of the easiest magnetic axis from one region of exchange coupled grains to the other as it has been recently observed for the field annealed FINEMET alloys [10].

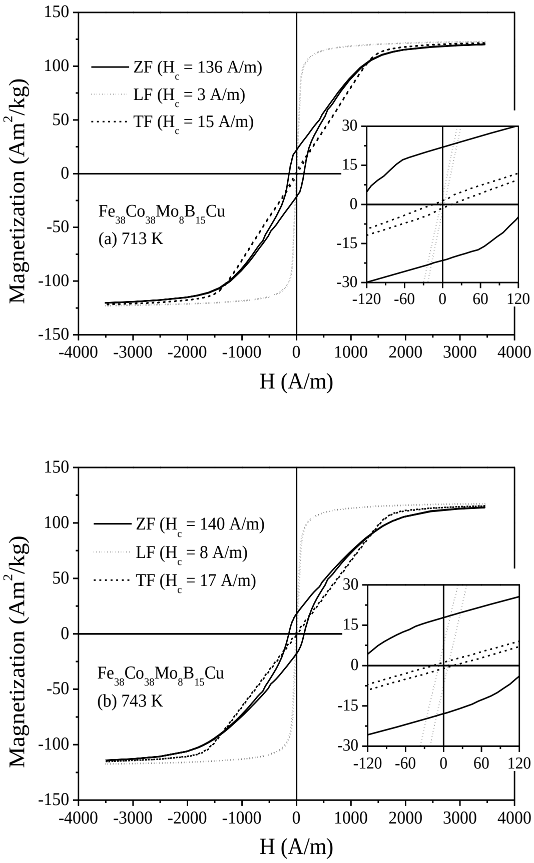

Figure 2. Hysteresis loops for $Fe_{38}Co_{38}Mo_8B_{15}Cu$ after different field annealing for 1 hour at indicated temperature.

The effects of annealing under presence of external magnetic field were studied also in the vacuum degassed non-oriented silicon steel sheets with the following chemical composition: C=0.008, Mn=0.22, Si=2.9, Cu=0.048, P=0.006, S=0.008, Al=0.%5 N=0.02, Ti=0.002 wt.%. Strips with thickness of 0.5 mm were taken from an industrial line after cold rolling with 80% reduction. The cold rolled specimens were subjected to laboratory recrystallization annealing at 1073 K for 10 min in dry hydrogen atmosphere. The steel laminations were subsequently subjected to additional laboratory cold rolling with 4 % reduction. Before the final heat treatment the specimens were cut into strips of 100 mm × 9 mm with the longest side parallel to the rolling direction.

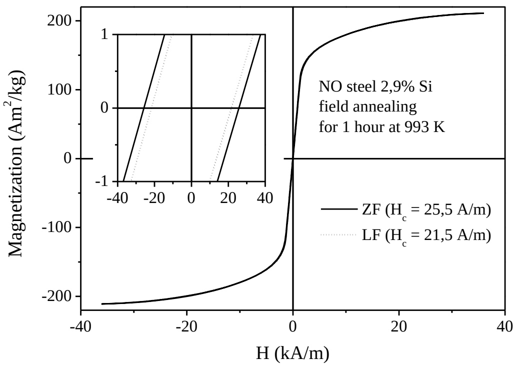

Figure 3. Hysteresis loops for non-oriented silicon steel samples after different field annealing for 1 hour at indicated temperature.

Finally, the rolled materials were annealed in pure hydrogen H_2 atmosphere upon dynamical heat treatment conditions. The annealing temperature applied to the experimental steel was 1223 K with holding time 180 sec. The basic idea behind this thermo-mechanical treatment was to promote the development of a coarse grained microstructure with pronounced intensity of cube and Goss texture components achieved during a final annealing [11,12]. Such microstructure leads to a significant decrease in the coercivity measured in DC magnetic field. Indeed, the low Hc values of ~ 23.5 A/m were obtained after the final heat treatment of our NO silicon steels strips, which were taken as the starting material for subsequent magnetic annealing experiments.

Field annealing (LF) was performed in the vacuum furnace inserted in the water cooled solenoidal coil that provided a magnetic field of 40 kA/m directed along the longest axis of the steel strip specimens. After annealing, the specimens were field-cooled down to room temperature with the cooling rate of 200 K/hour. The reference samples were annealed and cooled in a zero magnetic field (ZF). Fig. 3 shows the effect of magnetic field annealing on the coercivity of investigated non-oriented electrical steel sheets. The annealing temperature 993 K was selected to be well below the Curie temperature of the silicon steel containing 2.9 wt.% Si, so, the annealing was performed in the ferromagnetic state. Coercivity of LF-annealed samples slightly decreases as compared to the ZF-annealed sample, which indicates the beneficial effect of the field annealing. The improvement of the magnetic softness in the field annealed Fe(Si) solid solutions is related to the directional ordering of Si atoms in the Fe lattice [13].

4 CONCLUSION

The influence of the heat treatment under an external magnetic field on the magnetic properties of annealed material has been investigated in the $Fe_{38}Co_{38}Mo_8B_{15}Cu$ nanocrystalline alloys. We have shown that the crystallization of amorphous material in the longitudinal or transverse magnetic field is very powerful tool to tailor the shape of the hysteresis loops of these nanocrystalline alloys. Sheared loops with good field linearity and low coercive field were achieved after annealing in transverse magnetic field. A heat treatment of the samples under the presence of longitudinal magnetic field results in squared hysteresis loops characterized by the values of the coercive field in the range of 3 - 8 A/m. These H_c values are superior to those reported previously for HITPERM alloys. A marked response of the magnetic properties of FeCo-based nanocrystalline alloys to the magnetic field annealing can be utilized in their better adaptation to the potential electromagnetic applications. The magnetic field annealing effects were investigated also in the series of non-oriented electrical steel sheets containing about 2.9 wt.% Si. However, the contribution of induced anisotropy in NO silicon steels is rather small as it is superimposed by their non-negligible random magnetocrystalline anisotropy.

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