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Improvement of soft magnetic properties in $Fe_{38}Co_{38}Mo_8B_{15}Cu$ amorphous and nanocrystalline alloys by heat treatment in external magnetic field

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ABSTRACT

The influence of both longitudinal and transverse magnetic field applied during heat treatment on the soft magnetic behavior in the amorphous and nanocrystalline $Fe_{38}Co_{38}Mo_8B_{15}Cu$ alloys has been investigated. Sheared loops with good field linearity were achieved after annealing in transverse magnetic field. The value of induced anisotropy constant K_u after such annealing increased from 440 J/m^3 for thermally relaxed amorphous samples up to 935 J/m^3 for alloys in advanced stage of primary crystallization. The nanocrystallization under a presence of the longitudinal magnetic field results in squared hysteresis loops that are characterized by reduction of coercivity H_c to 3–7 A/m. Such low H_c values surpass those previously reported for FeCo-based nanocrystalline alloys.

1. Introduction

The nanocrystalline FeCo-based alloys prepared by devitrification amorphous precursors, called also HITPERM, display less favorable soft magnetic properties as compared to the nanocrystalline Fe-based FINEMET or NANOPERM alloys. However, they exhibit a higher saturation magnetic flux density and they are capable of operation at high temperatures [1,2]. In order to enhance the application potential of these alloys it is important to deepen knowledge about the available processing techniques that can be used to tailor and/or improve their soft magnetic properties. One possible way, which could be employed for this purpose, is thermal processing under the presence of external magnetic field, called also "magnetic annealing" [3].

The response of HITPERM alloys to the magnetic annealing has been recently studied by several groups [4–6]. The obtained results have clearly shown that the macroscopic uniaxial anisotropy K_u associated with annealing in the external magnetic field can dominate the very low effective random anisotropies in these materials. The shape of the hysteresis loop is governed by a balance between domain wall displacement and magnetic moment rotation processes. According to direction of the induced anisotropy, the magnetization curves with large or small squareness ratio could be obtained after annealing in longitudinal or transverse magnetic field, respectively.

Our previous study on the magnetic field annealing effects in the $(Fe_{1-x}Co_x)_{81}Nb_7B_{12}$ nanocrystalline alloys with various ratios of Fe/Co atoms has demonstrated that the improvement of the soft magnetic characteristics due to field annealing is most significant for the $Fe_{1-x}Co_x$ concentrations close to $x = 0.5$ [5]. Such behavior strongly indicates that the operative mechanism of induced anisotropy in these alloys is the magnetic atoms pair ordering [7,8].

In this work, a controllable field-induced magnetic anisotropy is produced in series of $Fe_{38}Co_{38}Mo_8B_{15}Cu$ thermally relaxed amorphous and partially crystallized samples. 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

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 h at different temperatures above the crystallization temperature in the presence of transverse (TF) or longitudinal (LF) magnetic field. In the case of TF-annealed samples, 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 the LF-annealed samples, the furnace was inserted into the water-cooled solenoidal coil that provided a magnetic field of 20 kA/m ori-

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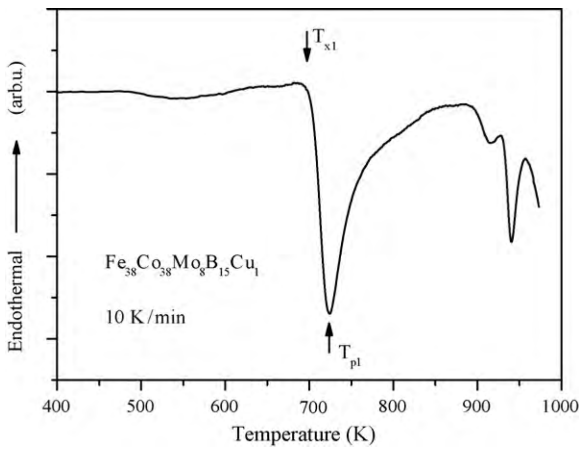

Fig. 1. DSC thermogram of amorphous $Fe_{38}Co_{38}Mo_8B_{15}Cu$ sample.

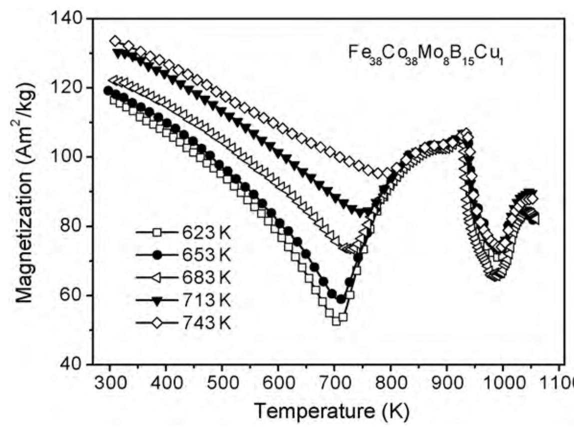

Fig. 3. Thermomagnetic plots of the thermally relaxed amorphous and nanocrystalline $Fe_{38}Co_{38}Mo_8B_{15}Cu$ samples.

ented 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 under the same conditions in a zero magnetic field (ZF).

The 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 magnetization measurements have been performed by vibrating sample magnetometer (VSM) over the temperature range from 300 K to 1100 K, in a field of 240 kA/m and with a heating rate 10 K/min. The soft magnetic behavior was investigated by using Forster type B-H loop tracer.

3. Results and discussion

Differential scanning calorimetry was used in order to get information on the crystallization behavior of the as-quenched amorphous alloy. Fig. 1 shows DSC thermogram measured in the temperature range where the crystallization takes place. The first exothermal peak with onset at T_{x1} = 695 K and the peak position at T_{p1} = 719 K corresponds to the formation of the nanocrystalline bcc-FeCo grains during the primary crystallization process. The crystallization of residual amorphous phase is associated with the subsequent peaks observed above 900 K.

The changes in microstructure upon annealing were examined by transmission electron microscopy. Fig. 2(a)–(d) shows TEM micrographs of differently heat treated samples. The sample annealed for 1 h at 623 K represents an example of thermally relaxed amorphous alloy. The sample annealed at 683 K for 1 h corresponds to the alloy with a low fraction of nanocrystalline grains. The presence of a higher amount of crystalline phase is clearly visible for the sample annealed at 713 K, while the sample annealed at 743 K for 1 h corresponds to the material in an advanced stage of the primary crystallization process. Typical dimensions of ultrafine crystalline grains determined from TEM micrographs range from 4 to 8 nm.

The temperature dependences of magnetization for the annealed amorphous and partially crystallized samples are shown in Fig. 3. The magnetization values of all samples show an initial monotonous decrease with the increase of measuring temperature. The partially crystallized samples exhibit higher magnetization values compared to the relaxed amorphous ones (annealed at 623 K

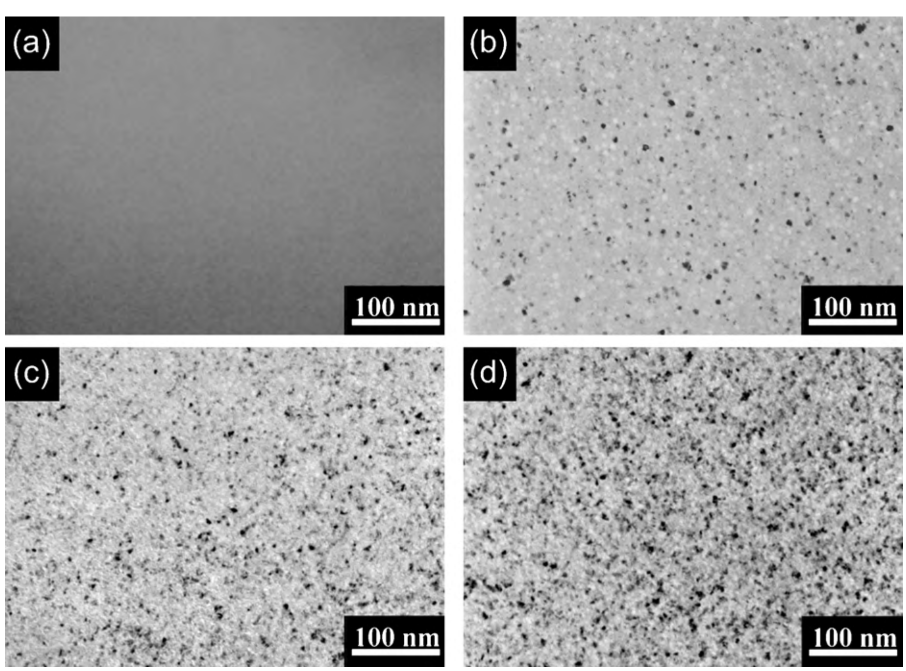

Fig. 2. TEM micrographs of the samples annealed for 1 h at 623 K (a), 683 K (b), 713 K (c) and at 743 K (d).

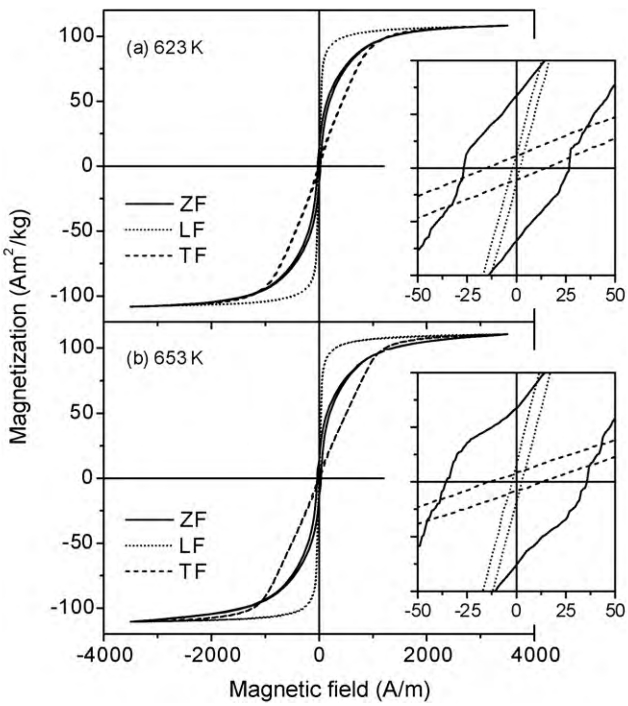

Fig. 4. (a and b) Hysteresis loops of thermally relaxed amorphous $Fe_{38}Co_{38}Mo_8B_{15}Cu$ after different field annealing for 1 h at indicated temperature.

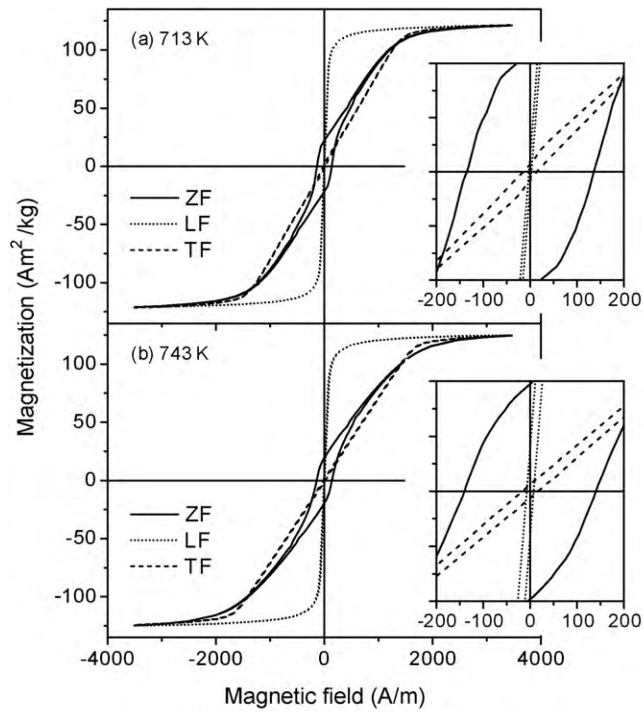

Fig. 5. (a and b) Hysteresis loops of nanocrystalline $Fe_{38}Co_{38}Mo_8B_{15}Cu$ after different field annealing for 1 h at indicated temperature.

and 653 K). It is attributed to the presence of bcc-FeCo crystalline phase with high magnetization and enhanced Curie temperature. After overcrossing the primary crystallization temperature, an increase in the magnetization is due to formation of additional crystalline particles. The character of the corresponding thermomagnetic plot for the sample annealed at 743 K indicates that the primary crystallization process is already in an advanced stage, while in the case of the sample annealed at 683 K a major part of transformation needs still to be accomplished. The sample annealed at 713 K corresponds to the material in a medium stage of primary crystallization process. These data are in good agreement with the previous TEM analysis.

The effect of field annealing on the hysteresis loops of the differently heat treated $Fe_{38}Co_{38}Mo_8B_{15}Cu$ samples is demonstrated in Figs. 4 and 5. Sheared loops with good field linearity were achieved for all alloys after annealing in transverse magnetic field. A heat treatment under the presence of longitudinal magnetic field results in squared hysteresis loops that are characterized by a significant reduction of the coercive field. The coercivity values for the ZF-, LF-, and TF-annealed samples are summarized in Fig. 6. The sample in the as-quenched amorphous state exhibits coercivity of 16 A/m. The ZF-annealing under zero field condition leads to marked increase of H_c values. Typical coercivities for the thermally relaxed ZF-annealed amorphous samples are around 30 A/m, while for the samples exhibiting an advanced degree of primary crystallization the coercive field approaches 150 A/m. From Fig. 6 it is evident that the field annealing substantially reduces the coercivity and the obtained H_c values after both LF- and TF-annealing do not show a striking difference between amorphous and crystallized samples. The improvement of the soft magnetic characteristics is more pronounced for the LF-annealed samples. The coercivity values after nanocrystallization are in the range of 3–7 A/m, i.e. they are superior to those previously reported for field annealed FeCo-based nanocrystalline materials. Fig. 6 also shows that the induced anisotropy constant K_u gradually increases with an increase of annealing temperature and its value reaches 935 J/m^3 for the sample annealed at 743 K. The lower K_u values as compared to the FeCoNbB [5,9] and FeCoZrB(Cu) [10] alloys are attributed to the lower saturation magnetization due to higher amount of non-magnetic elements in our sample. The saturation magnetization value for the conventional HITPERM-type FeCoZrB alloy can reach after advanced crystallization ~170 Am^2/kg [11], which is nearly 20% higher value as that reported for the present alloy.

The marked magnetic hardening after ZF-annealing seems to be connected with "self magnetic annealing" effects [3]. When aferromagnetic sample is annealed under the Curie temperature in demagnetized or non-saturated state, there will be an induced anisotropy parallel to the magnetization in the volume of the mag-

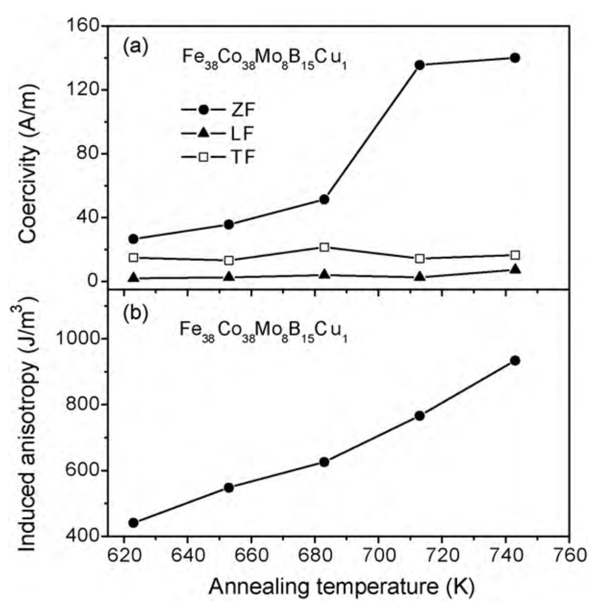

Fig. 6. The values of coercive field H_c (a) and induced anisotropy constant K_u (b) versus annealing temperature.

netic domains and induced helical anisotropy in the Bloch domain wall zones. The result of such annealing is that the domains and domain walls tend to be stabilized in the positions they occupied during the annealing. The domain wall stabilization effects result often in undesirable increase of coercive field and the corresponding hysteresis loops show a presence of steps due to depinnig of domain walls from the stabilized positions. Indeed, such steps can be seen from the enlarged central parts of hysteresis loops of thermally relaxed amorphous samples depicted in Fig. 4. In the case of nanocrystalline alloys the additional role can be played by the irregular magnetization patches that are fluctuating on the scale of a few micrometers as observed recently by digital enhanced Kerr-microscopy for soft magnetic nanocrystalline FINEMET alloys [12]. These fluctuations are most pronounced in the nanocrystalline samples with low induced anisotropy and they are virtually invisible in the amorphous samples. We assume that the stabilization of such irregular patches during the “self magnetic annealing” process leads to a further 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 a more simple domain configuration due to the uniform induced anisotropy, which in addition suppress the irregular magnetization patches connected with the angular dispersion of the easiest magnetic axis from one region of exchange coupled grains to the other as it was observed for the field annealed FINEMET [12].

4. Conclusions

In this work, a controllable field-induced magnetic anisotropy was produced in series of amorphous and nanocrystalline $Fe_{38}Co_{38}Mo_8B_{15}Cu$ samples with different amount of crystalline phase. We have shown that the heat treatment in the longitudinal or transverse magnetic field is very powerful tool to tailor the shape of the hysteresis loops of this material. The specimens annealed without the presence of external magnetic field show an appreciable increase of the coercivity with nanocrystallization. Sheared loops with good field linearity and the coercivity 13–21 A/m were achieved for the nanocrystalline samples after annealing in transverse magnetic field. The induced anisotropy constant K_u gradually increases with an increase of annealing temperature and its maximum value of induced anisotropy constant reaches the value 935 J/m^3. A heat treatment under the presence of longitudinal magnetic field results in squared hysteresis loops, which are accompanied by a strong reduction of the coercivity. The H_c values in the range of 3–7 A/m are to our knowledge a record mark for HITPERM-type nanocrystalline alloys.

Acknowledgments

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