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SHORT COMMUNICATION

ANNEALING INFLUENCE ON MAGNETIC BEHAVIOUR OF NANOCRYSTALLINE Fe-Hf-B BASED RIBBONS*

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Investigation of the influence of annealing type (furnace annealing, direct current (DC) Joule heating, furnace pre-annealing followed by DC Joule heating) on the magnetic behaviour of $Fe_{90}Hf_7B_3$ and $Fe_{88}Hf_7B_4Cu_1$ nanocrystalline ribbons obtained from amorphous precursor is presented in this paper. The obtained results show improved soft magnetic properties for the pre-annealed samples with respect to the others. The values of saturation induction and effective permeability for pre-annealed nanocrystalline samples are about 10% and 20%, respectively. These values are higher in comparison with those of the furnace annealed or DC Joule heated samples**.** The obtained results show the effectiveness of the two steps thermal treatment (furnace pre-annealing followed by DC Joule heating) in tailoring superior soft magnetic properties by strains relaxation, controlled nucleation and growth of crystalline grains.

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The development of nanophase materials with improved soft magnetic properties to be used as sensing elements is peculiarly important nowadays when more and more sensors are needed not only in the industry but also in our every day life. Fe-Hf-B nanocrystalline materials obtained by proper annealing from amorphous precursor are between the most attractive soft magnetic materials due to their excellent magnetic properties combined with the highest saturation induction Bs among the nanocrystalline soft magnetic materials [1,2].

Achievement of the demands on soft magnetic materials that are: high permeability (μ), low magnetostriction, high saturation induction (Bs), low hysterezis loss, high Curie temperature involves alloy chemistry and microstructure optimization that can be accomplished in the case of nanocrystalline materials obtained from amorphous precursor by means of proper annealing and by adding small amounts of special alloying elements.

Since much research time has already been devoted by various research groups to improve the soft magnetic properties of the Fe-based materials in nanocrystalline state using composition changes [3], it is the subject of the present work to investigate the influence of annealing type (furnace annealing, direct current (DC) Joule heating, furnace pre-annealing followed by DC Joule heating) on the magnetic properties of the Fe-Hf-B nanocrystalline ribbons obtained from amorphous precursor. The comparative study of the magnetic behavior of $Fe_{90}Hf_7B_3$ and $Fe_{88}Hf_7B_4Cu_1$ nanocrystalline ribbons obtained by the above mentioned annealing from amorphous precursor shows improved soft magnetic properties for the pre-annealed samples with respect to the others. As an example, the values of saturation induction and effective permeability for pre-annealed samples are about 10% and 20%, respectively, higher in comparison with those corresponding to the furnace annealed samples.

The $Fe_{90}Hf_7B_3$ and $Fe_{88}Hf_7B_4Cu_1$ alloy ingots were produced by arc melting pure elements in argon atmosphere. Rapidly solidified ribbons with cross-section of about 0.02×0.8 mm^2 were prepared by melt spinning on a fast rotating Cu-wheel with 450 mm diameter and a peripheral

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velocity of about 30 m/s, in argon atmosphere. In order to induce the nanocrystalline phase, the amorphous ribbons were subjected to three types of annealing: furnace annealing, DC Joule heating and furnace pre-annealing followed by DC Joule heating. The furnace annealing of the samples was performed in vacuum, for 1 hour, at temperatures in the range 500 - 650 °C. The DC Joule heating was performed by flowing an electrical current along the sample major axis, in vacuum. The furnace pre-annealing of samples was performed for 0.5 h, at 300 °C and 350 °C, where the crystallization does not take place yet. The pre-annealing was followed by DC Joule heating in vacuum. The amorphous structure of the as-quenched ribbons and the changes in samples microstructure upon annealing were monitored by X-ray diffraction (XRD). The XRD measurements were carried out using CoKα radiation. The influence of the annealing type on the magnetic properties of the studied samples was analyzed by room temperature magnetic measurements using a fluxmetric method. The saturation induction, B_s, was measured under applied field up to 10 kA/m. The effective permeability, μ_e, was measured at 50 Hz, under applied field up to 0.6 A/m. The saturation magnetostriction of the samples was determined by the small angle magnetization rotation (SMAR) method [4].

The optimum DC Joule heating for the achievement of good soft magnetic properties in $Fe_{90}Hf_7B_3$ nanocrystalline ribbons was determined to be for 0.57 A and 40 s annealing current and time, respectively. For $Fe_{88}Hf_7B_4Cu_1$ ribbons, the optimum DC Joule heating was achieved at 0.65 A annealing current, for 200 s. For 300 °C pre-annealed $Fe_{90}Hf_7B_3$ samples, the optimum DC Joule heating was found to be at 0.98 A and 900 s annealing current and time, respectively. The optimum DC Joule heating for 300 and 350 °C pre-annealed $Fe_{88}Hf_7B_4Cu_1$ ribbons was at 0.999 A for 3840 s and 2400 s, respectively.

Fig. 1 and 2 show the variation of X-ray diffraction patterns of $Fe_{90}Hf_7B_3$ and $Fe_{88}Hf_7B_4Cu_1$ ribbons, respectively, with the annealing type.

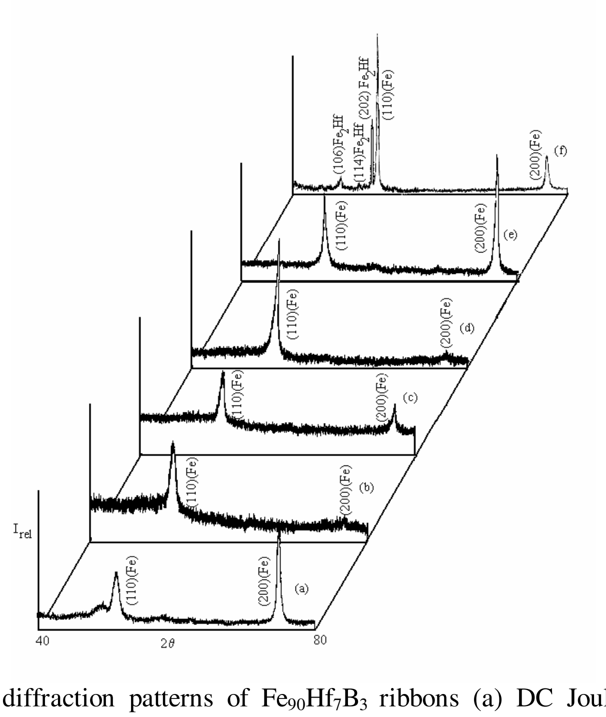

Fig. 1. X-ray diffraction patterns of $Fe_{90}Hf_7B_3$ ribbons (a) DC Joule heated; (b) furnace annealed at 500 °C, for 1 h; (c) furnace annealed at 550 °C, for 1 h; (d) furnace pre-annealed at 300 °C, for 0.5 h followed by DC Joule heating; (e) furnace annealed at 575 °C, for 1 h; (f) furnace annealed at 650 °C, for 1 h.

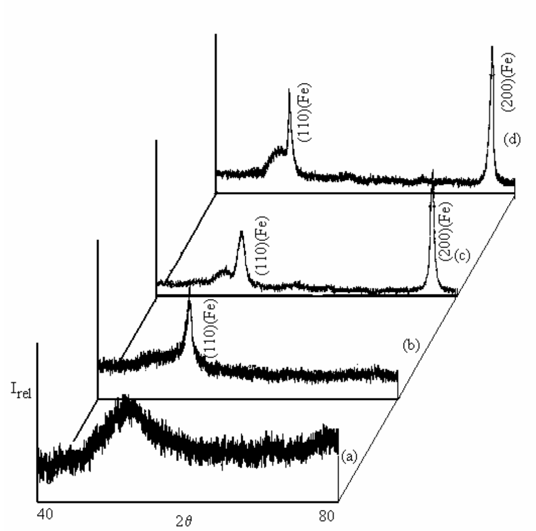

Fig. 2. X-ray diffraction patterns of $Fe_{88}Hf_7B_4Cu_1$ ribbons (a) as-cast; (b) furnace annealed at 550 °C, for 1 h; (c) furnace pre-annealed at 300 °C, for 0.5 h followed by DC Joule heating; (d) DC Joule heated.

In Table 1 are presented the values of grain size D, microstrain Δa/a (of II and III orders) and amorphous phase concentration c_a for as-cast and annealed $Fe_{90}Hf_7B_3$ and $Fe_{88}Hf_7B_4Cu_1$ ribbons determined from the X-ray patterns analyses.

Table 1. Grain size D, microstrain Δa/a and amorphous phase concentration c_a for as-cast and annealed $Fe_{90}Hf_7B_3$ and $Fe_{88}Hf_7B_4Cu_1$ ribbons.

Sample	Annealing	C_a (%)	D (nm)	Δa/a (10^{-3})
$Fe_{90}Hf_7B_3$	1 h/500 °C	73	15.2	0
$Fe_{90}Hf_7B_3$	1h/550 °C	57	14.9	0
$Fe_{90}Hf_7B_3$	1h/575 °C	48	15.5	0
$Fe_{90}Hf_7B_3$	1h/650 °C	32	16.4	6.4
$Fe_{90}Hf_7B_3$	0.5h/300 °C+DC Joule heating	62	13.4	0.73
$Fe_{90}Hf_7B_3$	DC Joule heating	67	15.7	0
$Fe_{88}Hf_7B_4Cu_1$	as-cast	96	-	0
$Fe_{88}Hf_7B_4Cu_1$	1h/550 °C	48	14	0
$Fe_{88}Hf_7B_4Cu_1$	0.5h/300 °C+DC Joule heating	68.7	10.3	9.42
$Fe_{88}Hf_7B_4Cu_1$	DC Joule heating	70.1	13.5	7.96

The grains size and microstrains were estimated by means of Delhez and collab. method [5]. As it can be seen, the smallest values of the grains size for both alloy compositions were achieved in the case of pre-annealed samples.

In Table 2 are presented the values of the effective permeability μ_e, saturation induction B_s and saturation magnetostriction λ_s for $Fe_{90}Hf_7B_3$ and $Fe_{88}Hf_7B_4Cu_1$ ribbons after annealing. The values of saturation induction and effective permeability for the $Fe_{90}Hf_7B_3$ and $Fe_{88}Hf_7B_4Cu_1$ nanocrystalline ribbons obtained by pre-annealing are about 10% and 20%, respectively higher comparing to those of the furnace annealed samples.

Table 2. Effective permeability μ_e, saturation induction B_s and saturation magnetostriction λ_s for $Fe_{90}Hf_7B_3$ and $Fe_{88}Hf_7B_4Cu_1$ ribbons after annealing.

Sample	Annealing	μ_e *	B_s (T)	λ_s (x10^{-6})
$Fe_{90}Hf_7B_3$	1 h/500 °C	9.7 K	1.42	0.43
$Fe_{90}Hf_7B_3$	1h/550 °C	24.7K	1.49	- 0.75
$Fe_{90}Hf_7B_3$	0.5h/300 °C + DC Joule heating	27.1 K	1.56	-
$Fe_{90}Hf_7B_3$	DC Joule heating	23.8 K	1.53	-
$Fe_{88}Hf_7B_4Cu_1$	1 h/500 °C	9.6 K	1.0	0.3
$Fe_{88}Hf_7B_4Cu_1$	1h/550 °C	21.7 K	1.35	-0.76
$Fe_{88}Hf_7B_4Cu_1$	1h/600 °C	20.7	1.42	-1.2
$Fe_{88}Hf_7B_4Cu_1$	0.5 h/300 °C + DC Joule heating	23.9	1.54	-
$Fe_{88}Hf_7B_4Cu_1$	0.5 h/350 °C + DC Joule heating	24.5	1.58	-
$Fe_{88}Hf_7B_4Cu_1$	DC Joule heating	22.3	1.52	-

* f = 50 Hz

Generally, there are no microstrains in the studied samples. They may be caused by vacancies, dislocations, etc or may appear at high annealing temperature, as can be seen in Table 1, when the dilatation coefficient of the constituent solid solutions are different and consequently, at room temperature, do appear distortions of the internal structure.

The results of the magnetic measurements are in good agreement with the structure configuration of the samples obtained from the analyses of the X-ray diffraction patterns.

The saturation magnetostriction for the $Fe_{90}Hf_7B_3$ ribbons decreases during nanocrystallization process from positive values to zero and then changes to negative values due to the balance between the contribution of the residual amorphous matrix (λ_s >0) and crystallites (λ_s <0).

By analyzing the results, it can be reported that addition of 1% Cu to Fe-Hf-B alloy leads to a nanocrystalline structure with improved soft magnetic properties determined by smaller grain size and almost the same values of Bs.

The improvement of the soft magnetic properties of the nanocrystalline studied samples by furnace pre-annealing followed by DC Joule heating can be explained due to internal stress relaxation, decrease of the residual intergranular amorphous phase and a more homogenous distribution of the α-Fe grain size.

The obtained experimental results and observations on the annealing type of the amorphous precursor for preparation of $Fe_{90}Hf_7B_3$ and $Fe_{88}Hf_7B_4Cu_1$ nanocrystalline ribbons show the effectiveness of the two steps thermal treatment (furnace pre-annealing followed by DC Joule heating) in tailoring superior soft magnetic properties by strain relaxation, controlled nucleation and growth of crystalline grains.

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