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The structure and magnetoelastic properties of the Fe-based amorphous alloy with Hf addition

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The paper presents results of investigation performed on newly developed Fe-based amorphous alloy with Hf addition. Structural, magnetic and magnetoelastic properties were tested. Results confirmed that addition of Hf decrease the stress sensitivity of the Fe-based alloy. For this reason $Fe_{85.4}Hf_{1.4}B_{13.2}$ alloy is significantly less stress sensitive than other Fe-based amorphous alloys.

1. INTRODUCTION

Recently, nanocrystalline materials which structure is formed in nanoscale have aroused researchers' interest. The nanocrystalline Fe-based alloys obtaining by controlling crystallization of amorphous alloys are a large group among this class of materials. The main parameters of two phase nanocrystalline ferromagnetic materials are: the grain size and volume fraction of the crystalline phase in structure after heat treatment of amorphous precursor. One of the conditions of forming nanocrystalline structure is separating temperatures range of nucleation and grain growth of the crystalline phase in amorphous matrix. However optimal soft magnetic properties are obtained by applying the heat treatment, defined as annealing at temperature T_{op}, which corresponds with maximum value of permeability [1,2].

Nanocrystalline soft magnetic alloys, from among the interesting group are Fe-M-B (M=Nb, Hf, Zr) ternary alloys called NANOPERM™ [1,3,4,5], show good properties combining high saturation induction (typical for Fe-based amorphous alloys) and vanishing magnetostriction, low coercive and high permeability (typical for Co-based amorphous alloys) [6,7,8-10].

The general understanding and controlling mechanisms determining the magnetic anisotropy, which influence on initial permeability (μ_i) and coercive force (H_c) are the most important to design soft magnetic materials.

Influence of mechanical stresses caused by external forces on magnetic properties of the alloy (so called magnetoelastic Villari effect) is very important from practical point of view. Due to high initial permeability amorphous and nanocrystalline alloys have low magnetocrystalline anisotropy. As a result participation of stress induced anisotropy energy in total free energy of such material is significantly higher than in a case of crystalline material [11]. Moreover magnetoelastic Villari effect is critical for miniaturized inductive components. In such case even small external forces may generate significant stresses in the core. For this reason properties of inductive components may be changed. Such external forces may be created in assembly of the component or as a result of thermal dilatation. Changes of the magnetic properties of the inductive component may produce significant changes in its functional parameters. Such changes may lead to the increasing of the losses in the core of the switching mode power supply. As a result such supply may work unstable or even may be thermally damaged. For this reason magnetoelastic properties of the amorphous and nanocrystalline materials should be taken into consideration in time of the development of the new amorphous or nanocrystalline alloy.

Significant stress sensitivity of the amorphous and nanocrystalline alloys creates possibility of utilizing this effect in construction of stress and force sensors. In a case of such sensor ferromagnetic material can be both sensing and construction element. In opposite to strain gauge sensors, in magnetolastic sensors, sensing element should not be mounted at elastic bar with adhesive layer. Moreover magnetoelastic stress and force sensors based on newly developed amorphous and nanocrystallie alloys (such as HITPERM) may operate in the high temperature range. Temperature of operation of such sensor is limited only by the Curie temperature. This temperature depends on alloy composition and can be about 600^0C for some alloys. As a result temperature of operation of magnetoelastic sensors can be significantly higher than strain-gauge sensors.

In the present work investigations of structure and influence of stress on magnetic properties of amorphous $Fe_{85.4}Hf_{1.4}B_{13.2}$ alloy after controlled crystallization has been undertaken.

2. EXPERIMENTAL PROCEDURE

The material was composed on the base of the ferromagnetic element (Fe - 85,4% at) what provides high saturation magnetic induction (B_s).
The rest elements: transition metal Hf (1.4% at.) and metaloid B (13.2 % at.) are added to promote glass formation in the precursor.

The Hf, recognized as a rare element, belong to the titanium group, plays a specific role in glass formation and in the nanocrystallization process. The Hf addition increases glass-forming ability in rapid solidification alloys. On the other hand the Hf addition in heat treatment process stabilizes amorphous phase range after part crystallization process [12]. Furthermore the growth of the primary Fe crystals is retarded in crystallization process due to the Hf localization (the grain boundary of the nanocrystalline αFe phase) and its atomic radius[12].

Experiments were performed on tapes of amorphous $Fe_{85.4}Hf_{1.4}B_{13.2}$ alloy obtained by planar flow casting method. Tapes had 0.024 mm thickness and 10 mm of width. Sections of tapes of 120 mm length were annealed in temperature range from 373÷1023 K in vacuum. The annealing time was constant and equal 1 h, with step of 50 K. The crystallization temperatures (T_{x1} and T_{x2}) of amorphous alloy were determined from isochronous curve ρ of samples, using the linear heating rate 0.007 K/s with measurement “in situ” [1].

In order to conduct structural study, the high-resolution electron microscope (HRTEM) JEM – 3010 in the range of $1.0 \cdot 10^5 \times$ to $1.5 \cdot 10^6 \times$ magnitude was used.

The measurements of initial permeability $\mu_i(T_a)$ (at H≈0.5 A/m and frequency f=1 kHz) in as quenched state and after annealing in temperature range 373÷1023 K by 1 h were performed with the use of automatic device for measurements magnetic permeability.

The samples after 1-h optimisation annealing temperature at T_{op} which correspons to the maximum value of initial permeability, were used for magnetoelastic properties investigations. Newly developed investigation method of magnetoelastic properties of the ring core was used. The idea of this method is presented in Fig. 1. Stresses were applied to the sample (outside dimension 20mm, inside dimension 15 mm) perpendicularly to the direction of the magnetizing field H_m [13].

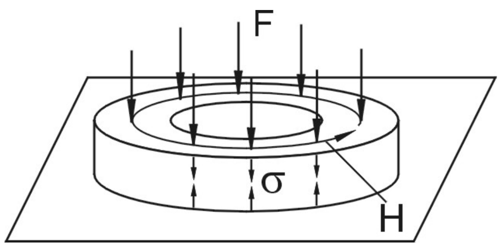

Figure 1. The general idea of applying uniform, compressive stress to the ring-shape sample

For practical realization of this idea of applying compressive stress to the ring sample nonmagnetic, cylindrical backings [14], presented in Fig. 2, were used. Special groves in the backings enabled sample to be winded. The compressive force was applied through the base backings.

The most important advantage of presented method is achieving uniform distribution of the both compressive stresses and magnetizing field in the ring-shape sample. Moreover both bulk and ribbon newly developed alloys may be tested. Due to the uniform distribution of stresses in the sample highly stressed area are absent. As a result magnetoelastic properties of even so brittle materials as nanocrystalline alloys may be investigated. [13].

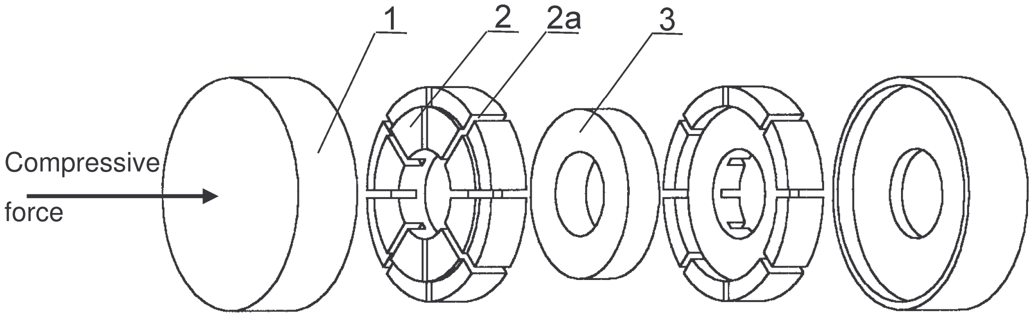

Fig. x3. Device for applying uniform, compressive stress to the ring-shape sample (patent pending P-345758) [x5] 1 – base backing, 2 – nonmagnetic, cylindrical backing 2a – grooves for the winding, 3 – sample under investigation

3. RESULTS AND DISCUSSION

The investigated $Fe_{85.4}Hf_{1.4}B_{13.2}$ alloy has amorphous structure in as quenched state – Fig. 3.

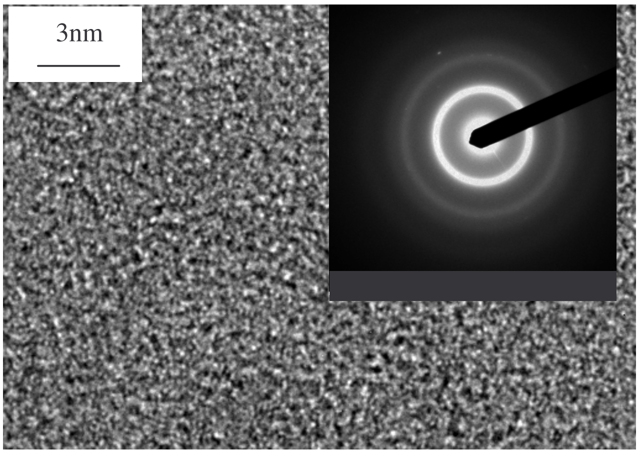

Figure 3. HRTEM micrograph for $Fe_{85.4}Hf_{1.4}B_{13.2}$ alloy in as quenched state

The obtained magnetic, i.e. initial permeability μ_i=151 (Fig. 4) and electric properties ρ=1,080 μΩm (Fig. 5) allow to classify $Fe_{85.4}Hf_{1.4}B_{13.2}$ alloy in as quenched state as a soft magnetic material.

The crystallization process of investigated alloy has two-stages character, i.e. T_{x1} =533 K (primary crystallization) and T_{x2}=703 K (polymorphous crystallization) - Fig. 5 and leads to changes of initial permeability (Fig. 4).

The highest value of initial permeability μ_i=239 (Fig. 4) have been achieved at annealing temperature 523 K denotes as temperature T_{op} of optimisation annealing treatment [1,2]. In the structure after annealing at T_{op} beside the amorphous phase the αFe crystalline phase of grains changing from 5÷25 nm appears (Fig. 6).

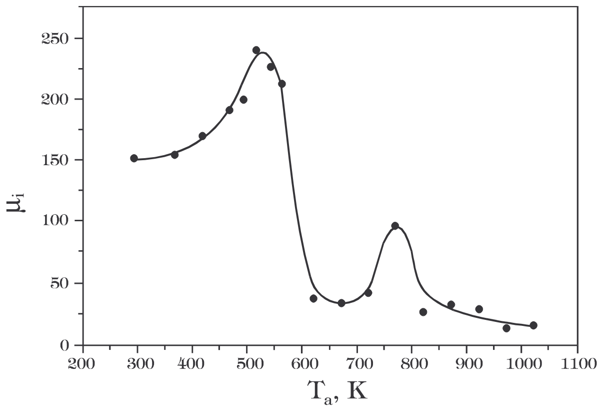

Figure 4. The initial magnetic permeability μ_i measured at room temperature for $Fe_{85.4}Hf_{1.4}B_{13.2}$ alloy after 1 h annealing at temperature T_a

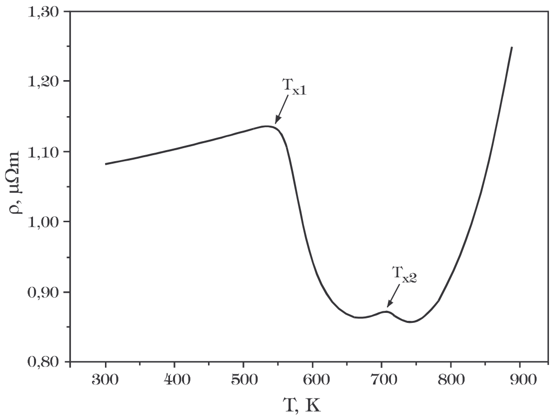

Figure 5. The isochronal resistivity curve of $Fe_{85.4}Hf_{1.4}B_{13.2}$ alloy determined for the heating rate 0.007 K/s

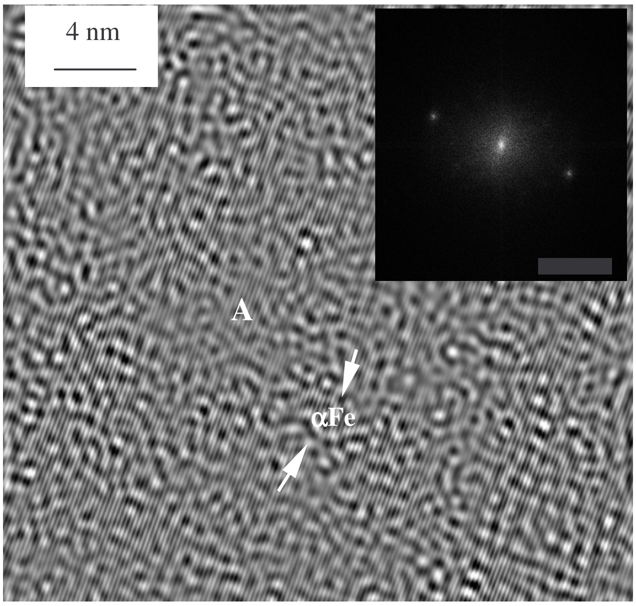

Figure 6. HRTEM micrograph for $Fe_{85.4}Hf_{1.4}B_{13.2}$ alloy after annealing at T_{op}=523 K by 1 h

The family of hysteresis loops for increasing value of the magnetizing field, for $Fe_{85.4}Hf_{1.4}B_{13.2}$ alloy after annealing in temperature 523 K for 1 hour, is presented in Fig. 6.

Due to the technical difficulty of measuring saturation flux density B_s the Jiles-Sablik model [15] of hysteresis loop was used. Parameters of the model were calculated with the Hook-Jewis optimisation method, by the criteria of the lower value of the sum of squares of the differences between the model and experimental results [16]. Finally, contribution of the variation described by the model in total variation of the experimentally achieved hysteresis loop (described by r^2 factor), for magnetizing field equal 3.7 kA/m, was equal 99.8 %. As a result of simulation the value of saturation flux density B_s was equal 1.58 T. Experimentally measured value of the coercive force H_c of the investigated alloy was equal 25 A/m (for magnetizing field H=1 kA/m).

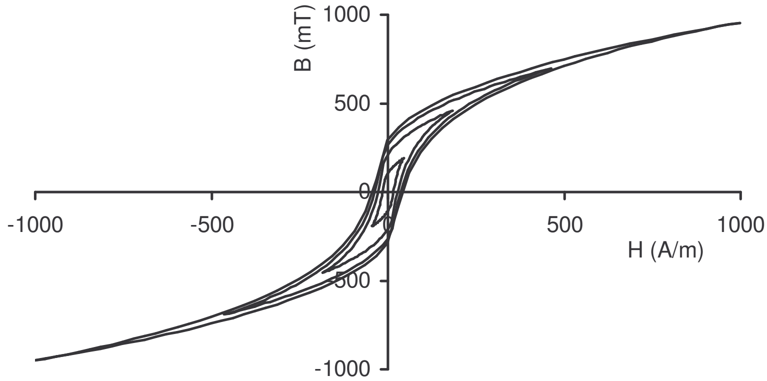

Figure 7. Familly of the hysteresis loops for $Fe_{85.4}Hf_{1.4}B_{13.2}$ alloy after annealing in temperature 523 K for 1 hour

When saturation flux density B_s of the $Fe_{85.4}Hf_{1.4}B_{13.2}$ alloy is known, value of the anisotropy <K> of the samples can be calculated. The assumption, that annealing does not change the value of saturation flux density B_s was made. Value of anisotropy <K> for both samples before and after annealing was calculated from the value of initial permeability μ_i, according to the equation (1) [17,18]:

$$<K>=\frac{{B_s}^2}{2\mu_0\mu_i} \tag{1}$$

Value of the initial permeability of the samples before and after annealing was equal as follow 151 i 239. As a result the value of anisotropy <K> was equal 6.5 i 4.1 kJ/m^3.

Influence of the mechanical, compressive stresses σ applied with the method presented in Fig. 1, are presented in Fig. 8.

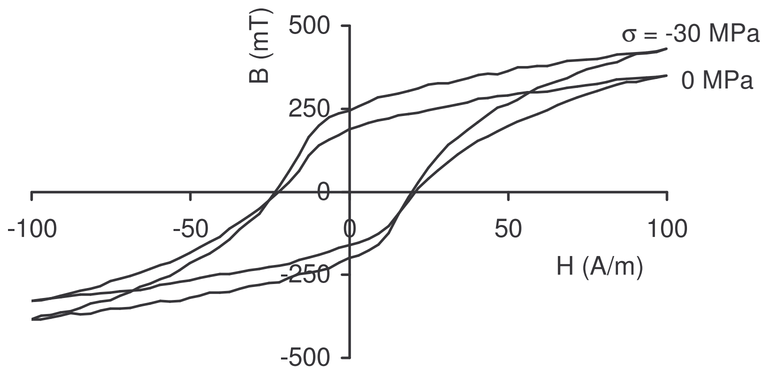

Figure 8. Influence of the mechanical, compressive stresses on hysteresis loop of the $Fe_{85.4}Hf_{1.4}B_{13.2}$ alloy after annealing in temperature 523 K for 1 hour

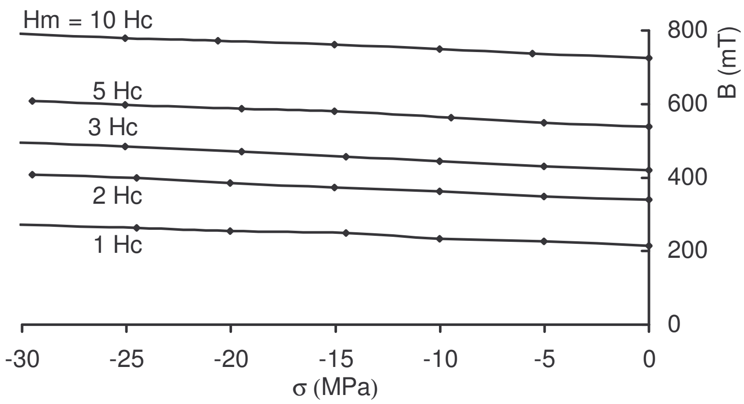

Figure 9. Changes of the flux density B in ring-shape core made of annealed $Fe_{85.4}Hf_{1.4}B_{13.2}$ amorphous alloy, under compressive stresses σ, for constant value of magnetizing field H_m (value of H_m is presented as the multiplied value of coercive force H_c)

Changes of the flux density B as a function of applied compressive stresses σ (for constant value of magnetizing field H_m) applied to the annealed core are presented in Fig. 9. Under influence of stresses up to 30 MPa value of the flux density B in core increases about 27%. Characteristics presented in Fig. 9 $B(\sigma)_H$ are approximately monotonous.

4. CONCLUSIONS

The worked out investigations have shown that amorphous $Fe_{85.4}Hf_{1.4}B_{13.2}$ alloy annealed in the temperature range 373÷1023 K is characterized by redistribution of the phases stages. The difference between temperature crystallization T_{x1} and T_{x2} is equal to 170 K. This effect is benefit from controlled crystallization process point of view The aim of the crystallization is forming two-phase structure consisting of αFe crystalline phase and amorphous matrix. It makes proceeding of nanocrystallization process easier and reducing the risk of forming the undesirable borides which cause the decrease of magnetic properties (μ_i) NANOPERM™ alloys [19].

The optimisation of soft magnetic properties is obtained with the use of controlled crystallization of amorphous alloys. This process can be explained by forming αFe crystalline phase in amorphous matrix and is connected with thermal activation. The structure of $Fe_{85.4}Hf_{1.4}B_{13.2}$ alloy after optimisation annealing at T_{op}=523 K, near to T_{x1} =533 K (primary crystallization temperature), is characterised by excellent soft magnetic properties (μ_i=239) and consists of αFe crystalline phase of grains changing from 5 to 25 nm and the amorphous matrix.

The influence of compressive stresses applied accordingly to presented method causes the increase of the value of flux density. On the basis of analysis of the total free energy of the material [20] it can be predicted, that value of the saturation magnetostriction of the $Fe_{85.4}Hf_{1.4}B_{13.2}$ alloy is positive. This prediction may be also justified by high percentage content of the iron in the alloy.

Because the Villari point was not reached ($B(\sigma)_H$ characteristics presented in Fig. 9 are monotonous) it can be assumed that investigated material has high residual stresses. Such stresses may be caused by presence of the Hf addition. Investigated material has significantly lower stress sensitivity than other iron-based amorphous alloy [20,21]. For this reason $Fe_{85.4}Hf_{1.4}B_{13.2}$ alloy may be utilized in construction of more stress-resistant inductive components.

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