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Annealing Temperature Affecting the Nanocrystalline Alloys' Physical Properties

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Abstract*— The effect of the annealing temperature on the mechanical and magnetic properties of the amorphous alloys based on ferrum and copper or without the latter has been studied. The coefficient of thermal expansion was found to change when the nanocrystalline alloy $Fe_{72,5}Cu_1Nb_2Mo_{1,5}Si_{14}B_9$ was being cooled at about t 350° C, which may be attributed to the ferromagnetic bond between nanograins. The copper content is shown to determine the varying nature of the amorphous alloy magnetic properties' dependence upon the annealing temperature.

Index Terms— **Annealing temperature, copper concentration, coercive force, diameter of remanent curvature, diameter of brittle fracture, ribbon length, relative magnetic permeability.**

I. INTRODUCTION

AMORPHOUS AND NANOCRYSTALLINE alloys have long been produced on a commercial scale [1]. In spite of this, they still attract particular interest as the subject of physical investigation. Being heated or cooled, alloys undergo various structural changes. The increase of the nanocrystalline annealing temperature is known to result in forming the nanocrystalline grains, their size changing, as well as the proportion of crystal and amorphous phases' volumes and the phase structure of an alloy change. In particular, with the annealing temperature being increased, nanocrystalline phase nuclei are formed in the alloy amorphous precursor, then nanograins grow; their size changes as well as the ratio of crystalline and nanocrystalline phases volumes and the alloy phase composition [2], [3**]**. Amorphous ribbons of different composition were annealed under tensile stress. This yielded a creep-induced magnetic anisotropy with an easy magnetic plane perpendicular to, or an easy axis parallel to the ribbon direction, depending on the alloy composition. X-ray diffraction experiments and simple thermal expansion measurements showed that the stress-annealed samples revealed a structural anisotropy which was released by post-annealing as a residual strain. This strain increases with the annealing stress and is therefore correlated with the induced magnetic anisotropy. The origin of this frozen-in strain is discussed in terms of structural heterogeneity in the strength of local atomic bonds. It is suggested that the induced magnetic anisotropy is related to the local magneto-elastic coupling in regions with strong bonding forces [4].

The structural anisotropy of Fe–Si–B–Nb–Cu nanocrystalline alloys annealed under tensile stress was studied by x-ray diffraction techniques with transmission geometry. A clear difference was observed in the peak positions of the Fe-Si crystals under two different conditions: with the diffraction vector parallel or perpendicular to the applied stress. The strains calculated from the anisotropy of the peak positions showed a linear response to the applied stress, independent of Si content, indicating that the observed structural anisotropy was due to the quenching of the elastic strain not in the directional order of the Fe-Si pair. The induced magnetic anisotropy energy is well explained by the residual strains and their magnetostriction [5].

Such structural changes may have an ambiguous effect on different physical properties of the material. Typically, magnetic permeability or a coercive force is used as magnetic parameters reflecting the change of the alloy structure [6], [7].

The alloy is to be heated to and held at a specific temperature before spinning starts. Amorphous ribbons are to be further annealing.

The temperature effect of $Fe_{72,5}Cu_1Nb_2Mo_{1,5}Si_{14}B_9$ nanocrystalline alloy annealing on the mechanical and magnetic properties has been investigated. Ferrum based amorphous alloys $Fe_{81}B_{13}Si_4C_2$ and $Fe_{77}Ni_1Si_9B_{13}$ with different copper content were compared.

II. EXPERIMENTAL

The amorphous ribbons were obtained by melt quenching on a rotating disk.

The amorphous ribbons samples of different chemical composition in the form of a flat ribbon 20-25 μm thick and 10 or 20 mm wide, wound as a torus with an outside diameter of 32 mm, the inside one of 20 mm and 10 mm high, were annealed at different temperatures in the air, being exposed to the specified temperature for 30 minutes.

A ribbon of 1000 mm long and 20 mm wide was being heated in the air by passing the electric current along the ribbon axis, and the ribbon temperature was measured by a thin thermocouple.

Static hysteresis loops, initial magnetic permeability $\mu_{0.08}$ and specific magnetic dissipation $P_{0.2/20}$, as well as magnetic hysteresis squareness ratio K_S were measured at annealing temperatures for 1 hour of ageing $P_{0.2/20}$ at frequency of 20 kHz and induction 0.2 T. Losses were estimated by means of dynamic hysteresis loops.

III. RESULTS AND DISCUSSION

Fig. 1 shows changing the ribbon length Δ, depending upon the heating temperature and the cooling temperature T_a.

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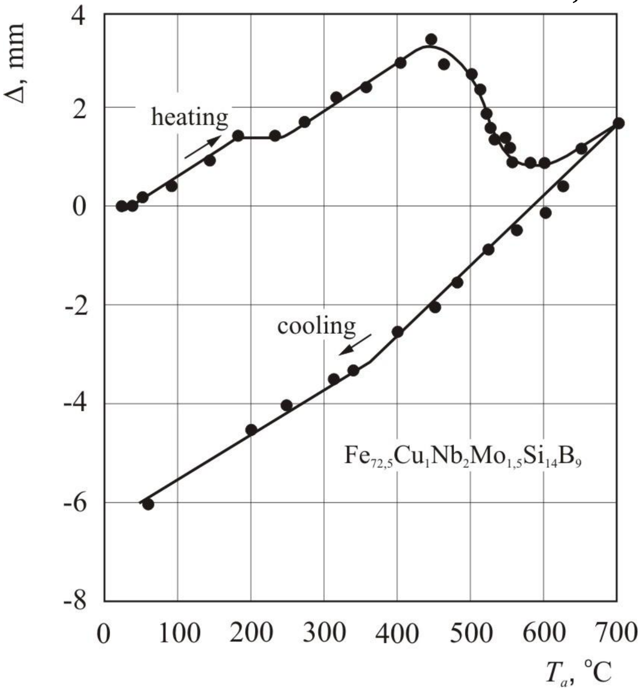

Fig. 1. Changing the ribbon length Δ made of $Fe_{72,5}Cu_1Mo_{1,5}Si_{14}B_9$ alloy, depending upon the heating temperature and the cooling temperature T_a.

In the process of heating up to 430° C, the ribbon length extends virtually linearly with a small platform over the area where the structural relaxation starts. This extension may be attributed to the thermal expansion of the material in the amorphous state [8].

With the further temperature increase approximately up to 560° C, the ribbon length is reduced monotonically. Evidently, this effect is related to the onset of the crystallization process. Crystallization being over, the ribbon starts extending again due to the thermal expansion of a new structure consisting of ferromagnetic nanocrystals of 10-20 nm introduced into the residual nonmagnetic matrix [9].

There are two linear portions on the cooling curve, with the change in the curve slope occurring approximately at 350° C. Changing the curve slope at this temperature can be attributed to the new magneto structural state of the material. The numerical value of 350° C agrees with the Curie temperature of the amorphous phase. With the temperature above this, ferromagnetic nanocrystals are separated from each other by an amorphous nonmagnetic matrix. If the temperature is lower, the amorphous matrix, becoming ferromagnetic, ensures a magnetic coupling of the nanocrystals and causes a new magneto structural state of the material, this state possessing a different coefficient of the material thermal expansion [10].

Crystallization being over, the ribbon starts extending again due to the thermal expansion of a new structure consisting of ferromagnetic nanocrystals, mainly α-$Fe_{80}Si_{20}$ size of 10-20 nm introduced into the residual amorphous nonmagnetic matrix [3], [9].

Fig. 2 shows the dependence of the diameter of remanent curvature D_r after annealing the ribbon of $Fe_{72,5}Cu_1Nb_2Mo_{1,5}Si_{14}B_9$ alloy at different temperatures on the mandrel of 32 mm diameter. It is seen that the remanent curvature is approaching the mandrel diameter after annealing at *t* 400° C. The numbers near the curve show the values of the initial magnetic permeability, measured at 1000 Hz. With the annealing temperature being increased, the initial magnetic permeability is regularly growing, first due to removing the internal hardening stresses in the ribbon, then due to forming the nanocrystalline structure.

The same figure represents the dependence of the mandrel diameter D_{bf}, on which the ribbon's brittle fracture occurs after thermal treatment at different temperatures. The ribbon brittleness increases as the temperature increases up to *t* 400° C, and then it remains constant in spite of the annealing temperature being increased. Then, at approximately *t* 520° C, the temperature which is normally taken as that of crystallization (the temperature of maximum heat production), the ribbon becomes more ductile.

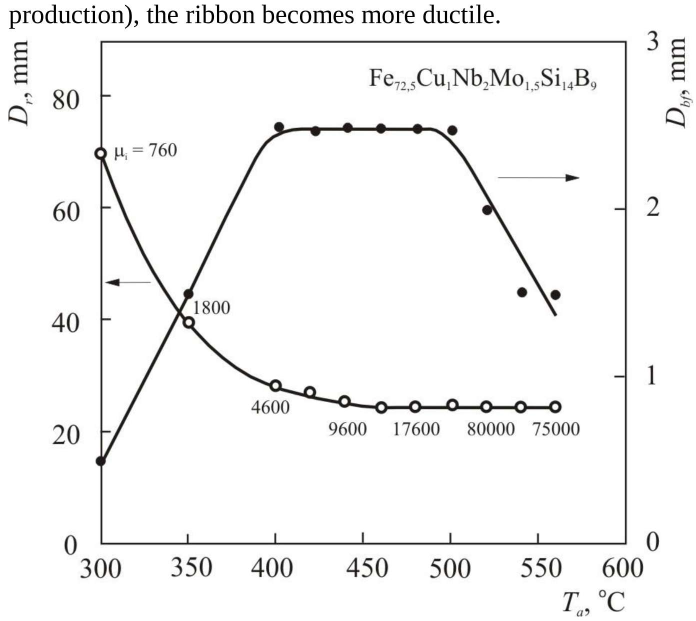

Fig. 2 Dependence of the diameter of remanent curvature D_r and the mandrel diameter D_{bf} on which the ribbon's brittle fracture occurs upon the annealing temperature of the $Fe_{72,5}Cu_1Nb_2Mo_{1,5}Si_{14}B_9$ alloy-made ribbon.

It is necessary to note that while studying the brittleness of the ribbon made of $Fe_{72,5}Cu_1Nb_2Mo_{1,5}Si_{14}B_9$ alloy, no differences in the numerical values of the fracture diameter D_{bf} were found in bending the ribbon with its wheel side surface or free side surface outward, or with different states of the sample's edge. This property makes the nanocrystalline alloy very different from the amorphous one based on ferrum.

Fig. 3 represents the dependence of the cylindrical mandrel diameter D_{bf}, on which the $Fe_{81}B_{13}Si_4C_2$ amorphous ribbon's brittle fracture occurs, upon the annealing temperature T_a. The tests were carried out bending the ribbon with its wheel side surface or free side surface outward, i.e. in bending, the wheel side surface or free side surface were subjected to stretching. The edge, where cracking occurred, was formed by diamond wheel cutting. From Fig. 3 it follows that in bending tests the samples annealed within the 430-460° C temperature range are most brittle. A rather wide scatter of the points tested prevents from distinguishing the difference between the wheel side surface and free side surface. This scatter seems to be attributed to the different edge states, on which uncontrolled cracks originate.

To make the edge state homogeneous, the samples to be used for mechanical tests were subjected to primary breaking as a result of which the test sample edges were formed. After this treatment, the lines of the primary brittle fracture became the sample edges for the mechanical tests. The results of the mechanical tests are given in Fig. 3b. First, it is seen that the numerical values of the fracture diameter have remarkably decreased as well as and the test points scatter has reduced as compared to what is given in Fig.3a. Secondly, the difference in mechanical properties of the amorphous ribbon in bending with its wheel side surface or free side surface outward became evident. In the process, the wheel side surface turned out to be more brittle, and the curve of the fracture diameter dependence shows an abrupt jump at *t* 400° C.

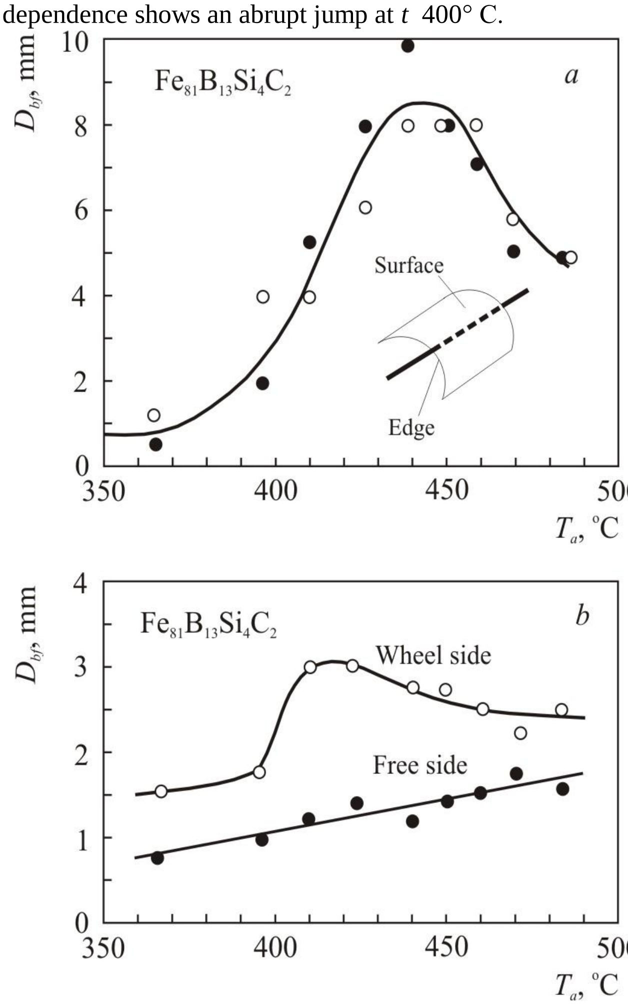

Fig. 3 Dependence of the mandrel diameter D_{bf} on which the ribbon's brittle fracture occurs after bending the free side surface (•) the wheel side surface (o) outward upon the annealing temperature T_a. The edge was formed by diamond wheel cutting (a) and as a result of the primary brittle fracture (b).

The fracture diameter of the free side surface is linearly increasing as the annealing temperature increases. Crystallization of ferrum-based amorphous alloys is greatly influenced by copper that, when heated, is uniformly distributed in the space as clusters up to 5nm size [11]. In the absence of copper, the crystalline phase nuclei of about 0.1 μm size originate near the surface [12], this phase is of a greater density compared to the amorphous layer inside the ribbon producing compressing stresses in the alloy with a positive constant of magnetostriction. Magnetization is aligned by compressing stresses mostly at right angles (perpendicular) to the ribbon surface [13], and the magnetic hysteresis loops become flat with a low residual magnetic induction B_r.

In this case, there is a 440-470° C temperature range in the $Fe_{77}Ni_1Si_9B_{13}$ alloy with a low Cu = 0.1 content at.% (Fig. 4a), within this temperature range, the magnetic permeability is essentially linearly dependent upon the annealing temperature, which is attributed to the gradual building up of the crystals' surface layer thickness. Varying the annealing temperature and duration, it is possible to constantly achieve a specified value of the magnetic permeability.

With a higher copper content 0.35 at.% (Fig. 4a), the temperature interval of the transition to a low magnetic permeability becomes very narrow.

The rectangularity coefficient of the magnetic hysteresis loop B_r/B_s reaches the value of 0,01 (Fig. 4b) in the alloy with a low copper content. The low rectangularity coefficient is indicative of the transverse magnetic anisotropy forming in the amorphous matrix. The complete crystallization being over, the rectangularity coefficient rises up to 0.7, that is close to the theoretical value of 0.832 for isotropically distributed non-interacting cubic grains [14]. High copper content alloys do not have low rectangularity coefficient values, and after complete crystallization, the rectangularity coefficient tends to be 0.8.

Different shapes of magnetic hysteresis loops are indicative of the varying character of the crystallization process flowing in the samples with different copper content [15]. Copper being absent at the initial stage, the surface crystallization with α-ferrum crystal nucleation dominates, the nuclei being mostly concentrated near the wheel side surface of the amorphous ribbon. In the process of annealing, copper forms clusters of several nanometers size all over the material volume, around which α-ferrum nuclei are formed, which aids in the crystallization process beginning at lower temperatures simultaneously all over the material.

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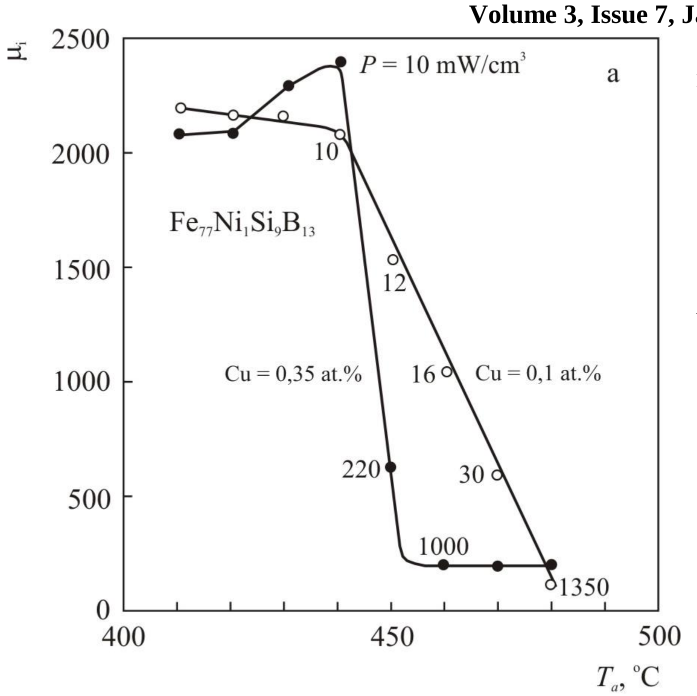

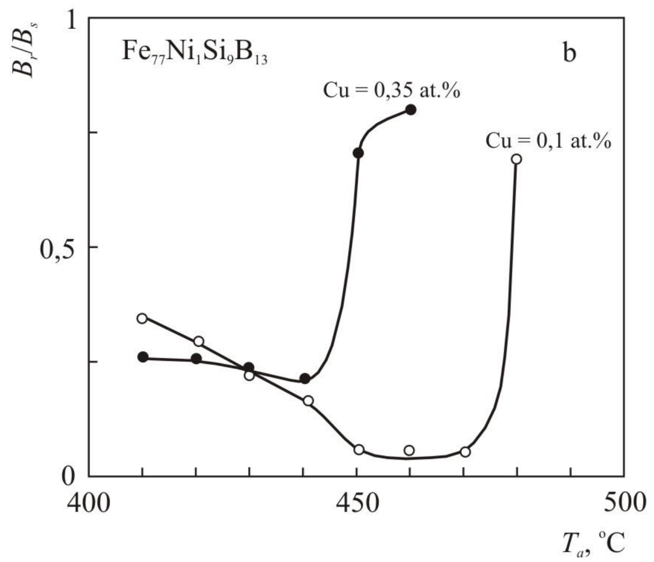

Fig. 4 Dependence of the initial magnetic permeability μ_i (a) and the rectangularity coefficient of the magnetic hysteresis loop B_r/B_s (b) upon the annealing temperature T_a of the $Fe_{77}Ni_1Si_9B_{13}$ alloy amorphous ribbon with different copper content 0.1 и 0.35 at.%. Figures close to the test points indicate the coercive force in A/м.

IV. Conclusion

The effect of the annealing temperature on the mechanical and magnetic properties of the amorphous alloys based on ferrum and copper or without the latter has been studied. The coefficient of thermal expansion was found to change when the nanocrystalline alloy $Fe_{72,5}Cu_1Nb_2Mo_{1,5}Si_{14}B_9$ was being cooled at about *t* 350° C, which may be attributed to the ferromagnetic bond between nanograins. The varying brittle nature of the amorphous precursor with copper ($Fe_{72,5}Cu_1Nb_2Mo_{1,5}Si_{14}B_9$ alloy) and without it ($Fe_{81}B_{13}Si_4C_2$ alloy) was found, the wheel side surface of the latter being more brittle, as compared to the free side one. The copper content is shown to determine the varying nature of the amorphous alloy magnetic properties' dependence upon the annealing temperature.

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2. Are the author more than 20 patents for inventions and useful models.

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2. Development and improvement of experimental methods of research of physical properties of metals and alloys

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