STRUCTURE OF THE AMORPHOUS FERROMAGNETIC MATERIAL Fe83,5 Cr2,5B14,3 AND ITS MAGNETIC PROPERTIES

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ABSTRACT

The article deals with the influence of changes of selected structural properties of amorphous ferromagnetic material Fe_{83,5}Cr_{2,5}B_{14,5} prepared by rapid quenching of the melt on its coercivity (as one of the most important magnetic parameters) during the process of hydrogenation and dehydrogenation.

Keywords: amorphous ferromagnetic material prepared by rapid quenching of the melt, clusterization, coercivity, hydrogenation, magnetoelastic anisotropy

1. INTRODUCTION

One of the major groups of materials for electronics purposes is the group of soft magnetic materials, whose main representatives are transformer plates. The first amorphous ferromagnetic materials were made a few decades ago and their magnetic properties are significantly better than those of transformer plates (e.g. coercivity is of one order lower). Therefore it is useful to know the relationship of structural properties of amorphous materials and their magnetic properties (e.g. coercivity and hysteresis losses). Therefore, amorphous materials in the form of a tape are increasingly used to produce toroidal transformer cores. Materials based on amorphous Fe-B are widely used and chromium is added for corrosion resistance. To ensure the stability of magnetic properties in operating conditions, it is useful to recognize the linkages between the changes in structural properties of amorphous materials and their magnetic property changes.

The dependence of the magnetic polarization J (or magnetic induction B) on the magnetic field strength His known as magnetization curve, or hysteresis loop (Fig. 1).



Fig. 1 Initial magnetization curve, hysteresis loop

We can determine most of the parameters which characterize ferromagnetic materials (saturation polarization J_S , coercivity H_C , remanence J_R , initial and maximum permeability, constant of total magnetic anisotropy, magnetization losses) from magnetization curves. The parameters that characterize ferromagnetic material from the perspective of possible technical applications are closely related to its physical state and structure. From this point of view, coercivity was chosen as a magnetic variable, which can characterize the structure of amorphous ferromagnetic materials.

There are two structural properties of this group of materials which have strong influence on magnetic parameters - a high level of internal stresses which are introduced into material during its preparation and the creation of atomic clusters (clusterization) which are generated by diffusion of atoms of glass-forming elements (boron) that can be looked upon as preparation for crystallization of metastable state. The absence of crystallographic anisotropy in amorphous materials brings about that magnetoelastic anisotropy is dominant.

2. METHODOLOGY OF THE MEASUREMENT

The studied material Fe_{83,2}Cr_{2,5}B_{14,3} was prepared by rapid quenching of the melt, in the form of ribbons, in the KFKI MTA in Budapest. Their amorphousness was checked by XRD and composition was determined by chemical analysis. Parameters of the sample are listed in the Table 1.

 Table 1
 Selected parameters of the sample

Sample	$J_{s}(T)$	$\lambda_s.10^6$	$T_{C}(K)$ [1]
Fe _{83,2} Cr _{2,5} B _{14,3}	1,08	22	502

Magnetic parameters of the sample were determined by magnetometer working on the principle of measurement of dispersive fields of the sample [2].

Heat treatment of amorphous materials is often used in the study of their structure and its possible modification. The disadvantages of this treatment are irreversible changes in amorphous structure. Atomic hydrogen was introduced into the material electrolytically because this process induces predominantly reversible changes in amorphous structure. Monitoring process of

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hydrogenation and consequential dehydrogenation contributes to the knowledge of stability of amorphous materials in the operating conditions. In the electrolytic hydrogenation 1N solution of H₂SO₄+10mg/l thiourea was used, electric current density was $5mA/cm^2$ and the time of the hydrogenation was 120 min.

In Fig. 2 there are shown the amount of hydrogen (in ppm) injected into the sample by hydrogenation, and the amounts of hydrogen in the sample during spontaneous dehydrogenation determined by the measurement of the amount of hydrogen released from the sample during dehydrogenation. The sample was in a vacuum chamber [3].



Fig. 2 Amount of the hydrogen in the sample during dehydrogenation

The experiment consisted of the determination of the studied parameters from the hysteresis loop measured on non-hydrogenated sample, then the sample was hydrogenated and hysteresis loops were measured on this sample at regular 30 minute intervals during spontaneous dehydrogenation.

3. RESULTS AND DISCUSSION

Fig. 3 depicts the course of coercivity (H_c) during hydrogenation and spontaneous dehydrogenation of Fe_{83,2}Cr_{2,5}B_{14,3} amorphous ribbon. By hydrogenation the H_c value increases by approx. 200%. At dehydrogenation a sharp decrease is observed. After several hours, H_c stabilizes at a higher value than before hydrogenation. It was expected that by introduction of about 210 ppm of hydrogen into the sample the level of internal stress would increase and may also affect the size of clusters, but no such a large increase of H_c was expected. We will try to explain it.

Disorders in the structure of real crystalline materials are generally the cause of irreversible displacement of domain walls. They include dislocations, inclusions (nonferromagnetic or with different magnetic anisotropy), grain boundaries and also influence of internal mechanical stresses. However, amorphous materials have neither crystallographic lattice with lattice disorders nor crystallographic anisotropy and there are no grains with boundaries in their structure.

Generally, coercivity can be expressed by the following formula

$$H_c = p.\frac{K_{eff}}{J_s} \tag{1}$$

where *p* is dimensionless material parameter (influence of structural disorders), K_{eff} is effective anisotropy and J_s is magnetic polarization of saturation.



Fig. 3 The course of coercivity during dehydrogenation

Magnetic polarization of the studied material J_s is known and during hydrogenation and dehydrogenation it changes only negligibly. The total (effective) anisotropy and magnetoelastic anisotropy can be determined from magnetisation curves. The courses of their changes are shown in Fig. 4.



Fig. 4 Courses of total and magnetoelastic anisotropy during dehydrogenation

It is clear that the change of effective anisotropy of the sample $Fe_{83,2}Cr_{2,5}B_{14,3}$ during hydrogenation and dehydrogenation is significant (40%), but the change of coercivity is several times higher. Therefore we have to find another mechanism for the increase of coercivity.

Amorphous materials prepared by rapid quenching of the melt do not posses lattice disorders although there is high level of internal stresses in their structure. In this case we can apply Kondorsky's stress theory of coercivity, which was developed for the case when the primary cause of magnetic hysteresis in ferromagnetic material is internal stress (it was originally developed for pure metals and alloys containing small homogeneous inclusions) [4]. According to this theory, during irreversible domain wall displacement the energy of ferromagnetic material changes due to the fact that the energy of a domain wall depends on its position.

Then coercivity can be expressed by the formula:

$$H_C = p_{\sigma} \cdot \frac{3}{2} \cdot \frac{\overline{|\sigma|} \lambda_S}{J_S} = p_{\sigma} \cdot \frac{K_{\sigma}}{J_S}, \qquad (2)$$

where $|\overline{\sigma}|$ is a mean value of internal stress, p_{σ} is a factor of dispersion of non-homogeneities of internal stresses and K_{σ} is magneto-elastic anisotropy. p_{σ} is determined as

$$p_{\sigma} = 3 \left(\frac{\delta_W}{l_{nc}} \right) / \left[1 + 3 \left(\frac{\delta_W}{l_{nc}} \right)^2 \right], \tag{3}$$

where δ_W is the thickness of domain walls and l_{nc} is a mean value of stress fields.

As can be seen in Fig. 5 coercivity depends on the ratio between the thickness of domain walls and mediumsized stress areas, calculated using the value of stress anisotropy and magnetic polarization of the sample in initial state.



Fig. 5 Dependence of coercivity and ratio δ_W / l_{nc}

It is clear from the picture that the ratio δ_W / l_{nc} significantly affects the size of coercivity. Coercivity is the greatest when the thickness of domain walls and medium-sized stress fields are approximately equal to each other.

From the equations (2) and (3) we can derive

$$l_{nc} = \frac{\delta_W}{\frac{K_{\sigma}}{2.H_C.J_S} \pm \sqrt{\left(\frac{K_{\sigma}}{2.H_C.J_S}\right)^2 - \frac{1}{3}}}.$$
 (4)

In amorphous materials prepared by rapid quenching of the melt the situation is more complex, because we must also take into account the change of the thickness of domain walls. Thus the thickness of domain walls can be another parameter affecting the magnitude of coercivity through the factor of dispersion of non-homogeneities of internal stresses. For the thickness of domain walls holds

$$\delta_W = \sqrt{\frac{AS^2 \psi^2}{(K_1 + 1.5\lambda_{100}\sigma)a}}, \qquad (5)$$

- where *S* is spin orientation (for ferromagnetic materials *S* = 1),
- ψ is the angle between two directions of spontaneous magnetization in adjacent domains $(\psi = \pi)$,
- A is exchange integral, for which it holds $A = \frac{3.k.T_C}{2.z.S.(S+1)} = 0.1kT_C$, where k is Boltzmann's constant (k = 1.38.10⁻²³ JK⁻¹), T_C is Curie temperature of the sample and z is the number of the nearest neighbor atoms of ferromagnetic atom (given that the density of studied material does not change and its value $\rho = 7, 6.10^{-3} kg.m^{-3}$ is slightly less than in $\alpha - Fe$, we chose z = 8),
- expression $(K_1 + 1.5\lambda_{100}\sigma)$ consists of expressions for the crystallographic and magnetoelastic anisotropy. For amorphous ferromagnetic materials without crystalline structure this expression is reduced to magnetoelastic anisotropy K_{σ} that is dominant in amorphous ferromagnetic materials prepared by rapid quenching of the melt,
- *a* is a lattice constant, for crystalline iron $a = 2.86.10^{-10} m$, for the studied materials it is a mean distance between atoms; since the densities of crystalline iron and studied material are very close, we use this constant as the mean distance between atoms in studied material.

From the above-mentioned considerations results that the expression (5) for amorphous materials in accordance with [6] can be modified as follows

$$\delta_W = \pi \sqrt{\frac{A}{K_{\sigma}.a}} = \pi \sqrt{\frac{0.1 \, k.T_C}{K_{\sigma}.a}} \,. \tag{6}$$

The thickness of 180° domain wall of the sample during hydrogenation and dehydrogenation was determined using (6). Its course is shown in Fig. 6.



Fig. 6 The thickness of 180° domain wall during hydrogenation and dehydrogenation

We can see that the thickness of 180° domain walls in amorphous material is several times greater than the thickness of domain walls in crystalline iron, where it less than about 30nm [5]. It is interesting, that the thickness of domain walls in amorphous material is not constant during hydrogenation and dehydrogenation, but an increase of internal stress causes reduction of its thickness.

Another structural feature affecting coercivity is the size of atomic clusters that is related to the size of stress centers. The mean size of stress centers was determined from (4) and it is shown in Fig. 7.



Fig. 7 Changes of the mean size of stress centers during hydrogenation and dehydrogenation

The course of the size of mean-sized stress centers shows its increase until hydrogenation is finished. If we assume that hydrogen enters amorphous matrix first, its expansion can result also in clusters expansion, and unequal stresses in the matrix and clusters also increase the size of stress centers.

In addition to the rate of internal stresses the ratio between the domain wall thickness and the size of stress centers is crucial for the behaviour of coercivity in amorphous materials prepared by rapid quenching of the melt. This means that changes of all mentioned variables may have a significant impact on coercivity.

4. RESULTS

Measurements of coercivity and decreasing branch of hysteresis loop from which the magnetoelastic anisotropy was determined are standard magnetic measurements, although the results of these measurements may lead to the knowledge of such structural features, such as the degree of internal stress, the size of stress centers that are closely related to the size of clusters, and also the thickness of domain walls. This allows more detailed description of processes in amorphous ferromagnetic material during hydrogenation and dehydrogenation:

During hydrogenation of amorphous material $Fe_{83,2}Cr_{2,5}B_{14,3}$ the mean value of internal stress increases that leads to the increase of a mean value of internal stresses as well as the increase of magnetoelastic anisotropy. This directly increases the coercivity (see (1)), and decreases domain walls thickness, which leads to an increase of coercivity too. The third mechanism which leads to the increase of coercivity is the enlargement of stress centers due to the non-homogeneous distribution of hydrogen atoms in the sample.

Hydrogen atoms leave a morphous sample ${\rm Fe}_{83,2}{\rm Cr}_{2,5}{\rm B}_{14,3}$ during dehydrogenation fast (cca 5 hours – see Fig. 2), but ongoing processes during dehydrogenation subside for 20 to 30 hours.

During dehydrogenation two parallel processes are in progress:

- release of hydrogen atoms induces successive relaxation of stress caused by the hydrogen introduced during hydrogenation,
- "lattice" distorted by the hydrogen facilitates diffusion of boron atoms at short distance, that results in :
 - the reduction of internal stress introduced into material during its preparation,
 - the increase of atomic clusters, acting as stress centers.

ACKNOWLEDGMENTS

This work has been supported by the Slovak Grant Agency VEGA 1/0136/10 and 1/0167/10.

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Received March 10, 2010, accepted July 12, 2010

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