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The Effect of Temperature and Frequency on Magnetic Properties of the $Fe_{81}B_{13}Si_4C_2$ Amorphous Alloy

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Abstract:

In this study it was investigated influence of temperature and frequency on permeability, coercivity and power loses of $Fe_{81}B_{13}Si_4C_2$ amorphous alloy. Magnetic permeability measurements performed in nonisothermal and isothermal conditions was confirmed that efficient structural relaxation was occurred at temperature of 663 K. This process was performed in two steps, the first one is kinetic and the second one is diffuse. Activation energies of these processes are: E_{a1} = 52.02 kJ/mol for kinetic and E_{a2} = 106.9 kJ/mol for diffuse. It was shown that after annealing at 663 K coercivity decrease about 30% and therefore substantial reduction in power loses was attained. Investigated amorphous alloy satisfied the criteria for signal processing devices that work in mean frequency domain.

Keywords: *Amorphous alloys; Structural relaxation; Annealing; Improving soft magnetic properties*

1. Introduction

The amorphous alloys (metallic glasses) are advanced functional materials due to a specific combination of properties. These materials are characterized by structure with absence of the distant order atom arrangement and characterized by high degree of anisotropy of physical properties [1-3].

The amorphous state of matter is, however, structurally and thermodynamically unstable and very susceptible to partial or complete crystallization during thermal treatment. The crucial limitation with respect to using metal glasses for high temperature applications arises from their restricted thermal stability. The onset of exothermic crystallization results in the formation of highly stable, but brittle intermetallic compounds. Further, for amorphous alloys that exhibit excellent magnetic properties the crystallization represents the limit at which these properties begin to deteriorate. For the case alloys that exhibit excellent magnetic properties in the two-phase nanocrystal-amorphous matrix structure, control of the crystallization kinetics allows the ability to tailor desired structure [4-6]. Therefore, the knowledge of alloys stability in a broad range of temperature due to different crystallization processes which occur during annealing is crucial [7].

Among these materials, the amorphous alloys of Fe and B have been very interesting because of the combination of soft ferromagnetic properties and high saturation magnetic flux

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density. These alloys are applicable in a variety of devices, such as transformers, magnetic sensors and recording heads [8-10]. The Curie temperature of these alloys increases slightly on replacement of boron by silicon. It was also concluded that the crystallization temperature increases with increase of content of silicon and decrease of content of iron and boron. Further, this results in a sharp increase of saturation magnetization extending from $Fe_{80}B_{20}$ to $Fe_{82}B_{12}Si_6$. It should be noted that the multicomponent alloys are easier to prepare in the amorphous state than the binary ones. Thus for the highest saturation magnetization alloy combined with ease of preparation, stability and lowest losses, the alloys of Fe-B-Si system are preferred [11].

Tailoring of functional properties of these alloys can be successfully performed over reduction of residual stresses during appropriate thermal treatments. Therefore, in this paper we study influence of structural relaxation on magnetic characteristics (coercivity H_c, relative magnetic permeability μ_r and power loses P_s) of amorphous alloy $Fe_{81}B_{13}Si_{14}C_2$. In order to estimate magnetic properties for applications in electrical engineering, B-H hysteresis loop measurements were made on the scale of frequencies from 50 Hz to 1 kHz and on different excitation magnetic field.

2. Experimental

Ribbon shaped samples of $Fe_{81}B_{13}Si_{14}C_2$ amorphous alloy were obtained using the standard procedure of rapid quenching of the melt on a rotating disc (melt-spinning). Thermal stability was investigated in a nitrogen atmosphere by the differential scanning calorimetry (DSC) method using SHIMADZU DSC-50 analyzer in the temperature region from room temperature to 900 K. Magnetic permeability measurements performed in isothermal conditions from 623 to 663 K during 20 minutes. B-H hysteresis loops were measured on toroidal core samples by Brockhaus MPG 100D measuring system.

3. Results and discussion

The differential scanning calorimetry method was used for investigating the crystallization process of the $Fe_{81}B_{13}Si_4C_2$ amorphous alloy. It was observed that the crystallization occurs from 783 to 823 K (see Fig.1). It has been established that the Curie temperature is about 693 K for amorphous state (that is also confirmed by DSC endo λ-peak position).

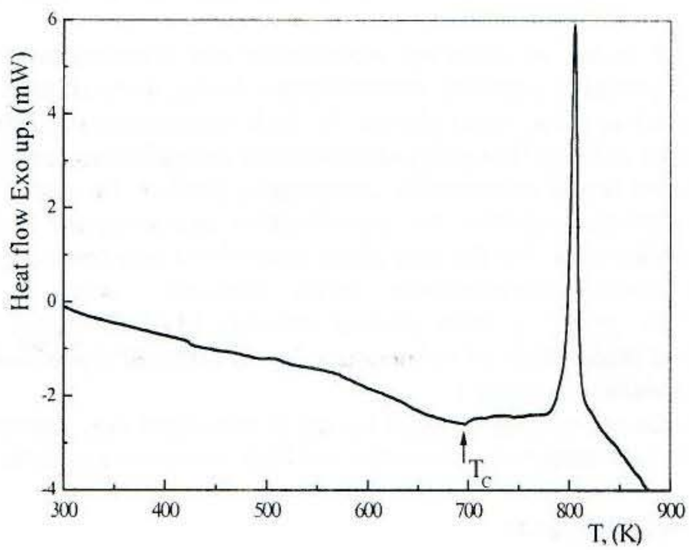

Fig. 1. DSC trace of $Fe_{81}B_{13}Si_4C_2$ amorphous alloy at heating rate of 20 K/min (arrow denotes endo λ-peak i.e. Curie temperature T_C = 693 K).

In order to prevent early stage of crystallization process experiment were performed in isothermal conditions from 623 to 663 K for 20 min. The diagram of the change of relative magnetic permeability presented in Fig. 2. confirms that structural relaxation was occurred. The analysis of these results reveal that the process was performed in two steps, the first one is kinetic and the second one is diffuse.

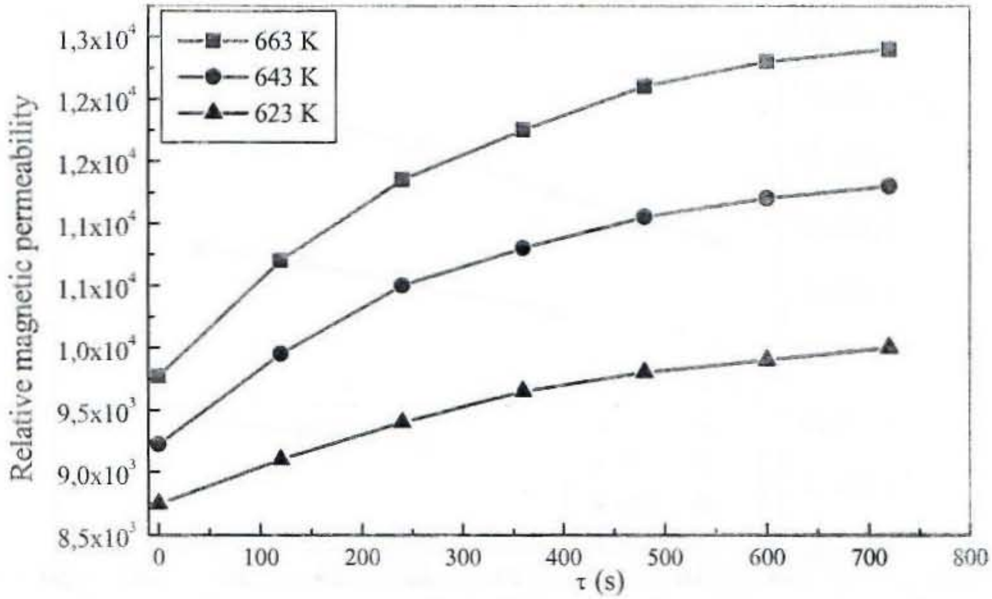

Fig. 2. The changes of relative magnetic permeability of $Fe_{81}B_{13}Si_4C_2$ amorphous alloy at three isothermal treatments 623 K, 643 K and 663 K.

The diagram in Fig. 3 shows logarithmic dependence of isothermal relative magnetic permeability on time $\ln \mu_r = f(\tau)$. In initial time one can see linear dependence associated to the beginning of structural relaxation that is activationally controlled by transitions of atoms from higher to lower energy state.

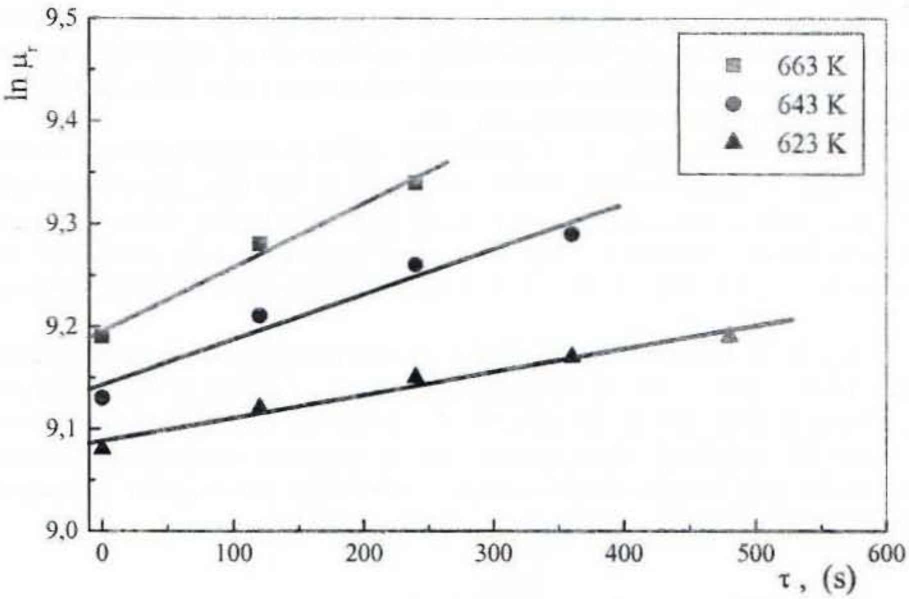

Fig.3. The changes of logarithm dependence of relative magnetic permeability at three isothermal treatments 623 K, 643 K and 663 K.

The second stage of the structural relaxation process is characterized by linear dependence $\mu_r = f(\tau^{1/2})$ as it is presented in Fig. 4. The observed dependence suggests that the second stage of the stress relief is a diffusion process due to the movement of the inter cavity atoms loading to the free volume decrease. Activation energies of these processes are: E_{a1}=52.02 kJ/mol for kinetic and E_{a2}=106.9 kJ/mol for diffuse.

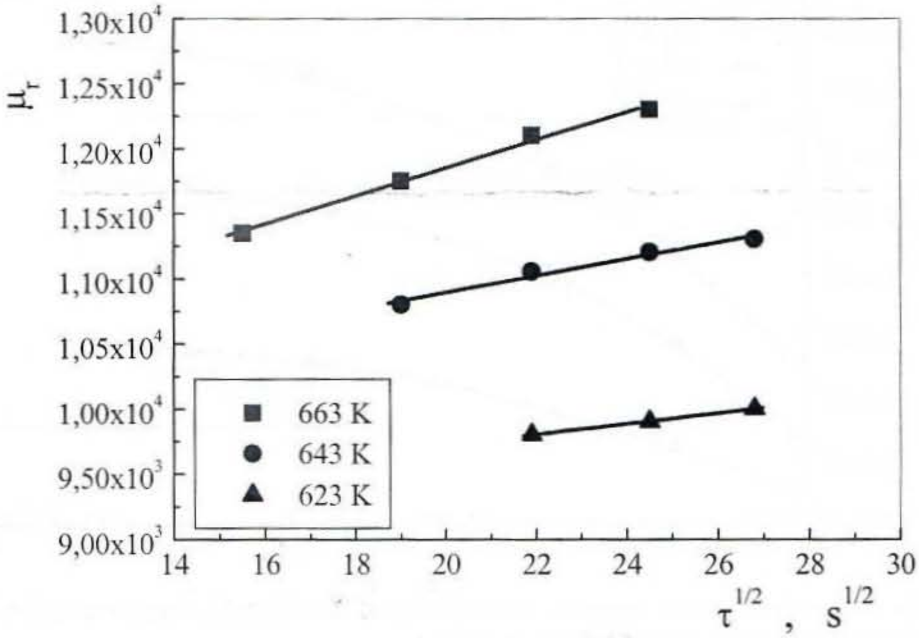

Fig. 4. The changes of relative magnetic permeability on square root of the process time at three isothermal treatments 623 K, 643 K and 663 K.

After these investigations it can be concluded that the most intensive structural relaxation were performed during annealing at temperature of 663 K. Therefore, one can expect that the magnetic characteristics of this sample are the most attractive. In a frame of the efficiently applications in electrical devices the main magnetic hysteresis properties such as coercivity, permeability and magnetic losses as a function of frequency were investigated. These parameters were measure of the magnetic softness and after their optimization material can be useful for electrical engineering purposes.

As it is shown on Fig. 5 after annealing at 663 K coercivity decrease about 30% at all frequencies and at magnetic fields intensities from 25 to 100 A/m. The increase of coercivity on magnetic field is specially observed at 50 A/m. For higher values of magnetic field coercivity is almost independent. The influence of frequency on the changes of coercivity of as-cast and annealed samples is the same, i.e. it can be observed the similar increase on curves in Fig. 5.

The role of magnetisation reversal is the real parameter of dynamic hysteresis loops. The effect of frequency with the sinusoidal flux density is shown in Fig. 6. The excitation in magnetic field amount had to 100 A/m, the frequency 50, 200, 400, 600, 800 and 1000 Hz. With increasing frequency, have greater rate of magnetic reversal, the hysteresis loops become broader and therefore the losses higher. This behaviour is caused by eddy-current and at higher frequencies additionally by spin relaxation processes.

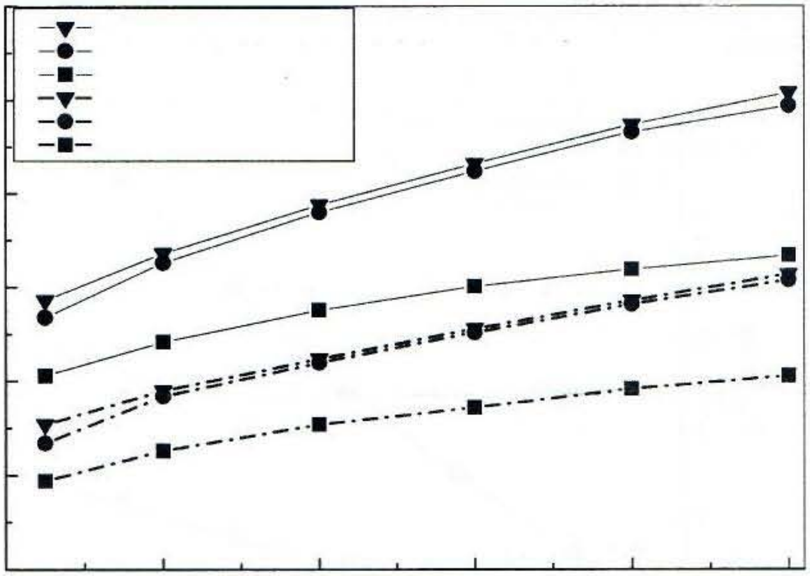

Fig. 5. The changes of coercivity of the $Fe_{81}B_{13}Si_4C_2$ at different frequencies after increase of maximum magnetic field H_{max} from 25 A/m to 100 A/m in as-cast state and after annealing at 663 K.

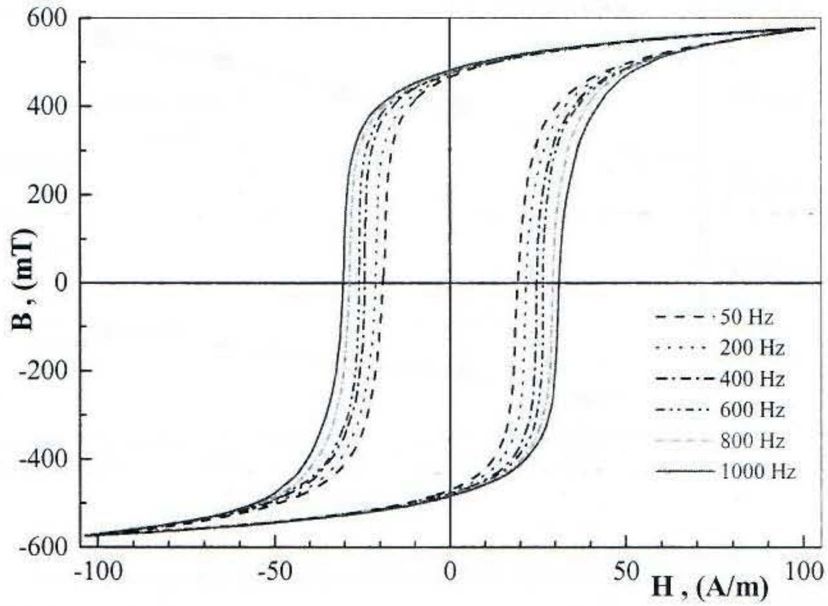

Fig. 6. Hystresis loops of the $Fe_{81}B_{13}Si_4C_2$ toroidal sample in as cast state at different frequencies (from 50 Hz to 1000 Hz).

It should be noted that the hysteresis loops are with high remanence ratio ($B_r/B_s \approx 0.8$) what can be very useful advantage of this magnetic material. The core losses that are proportional to the surface area of the hysteresis loop and consist of hysteresis and eddy-current losses. Frequency dependence of total power loss P_s referred to the core mass is shown in Fig. 7a. The sample annealed at 663 K exhibit strong reduction of P_S, i.e. after evolved stress relaxation the movement of the domain walls is easier and reduction of energy losses is attained.

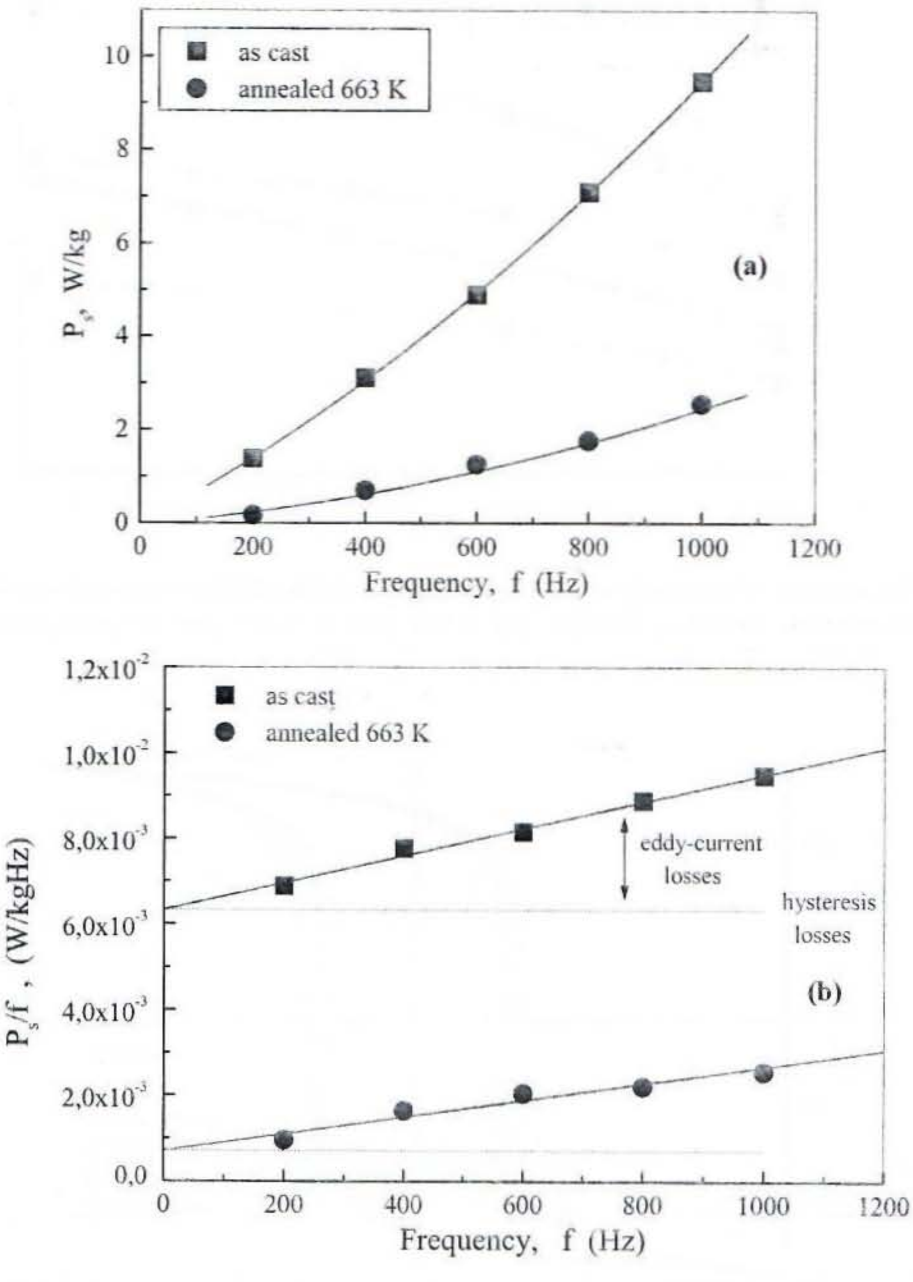

Fig. 7. a) Total power loose P_s referred to the core mass and b) the separation of magnetization reversal losses.

As the hysteresis losses are proportionally to the frequency (~ f) and eddy-current losses are proportionally to the square of frequency (~ f^2) it can be performed separation between these components. The results of separation of magnetization reversal losses of the $Fe_{81}B_{13}Si_4C_2$ toroidal sample are shown in Fig. 7b. Due to the high electrical resistivity and low thickness of the ribbon that is 35 μm (which both limit the eddy current losses) and due to the low hysteresis losses one can see low core losses. Obtained results of P_s = 3.25 W/kg for as cast and P_s = 0.67 W/kg for annealed sample (at 400 Hz; 0.57 T) are comparable with the data of P_s = 2 W/kg for METGLAS 2605 CO ($Fe_{67}Co_{18}B_{14}Si_1$ alloys at 400 Hz; 0.6 T) [12]. Therefore, investigated amorphous alloy satisfied the criteria for signal processing devices that work in mean frequency domain.

4. Conclusion

In this study it was investigated structural relaxation of $Fe_{81}B_{13}Si_{14}C_2$ amorphous ribbon and improvement of magnetic characteristics (coercivity H_c, relative magnetic permeability μ_r and power loses P_s). Performed analysis of isothermal relative magnetic permeability shows that the stress relief process was performed in two steps: the first one is kinetic with activation energy of E_{a1}=52.02 kJ/mol and the second one is diffuse with activation energy of E_{a2}=106.9 kJ/mol. The sample annealed at 663 K exhibit the highest level of stress relief and therefore the most attractive magnetic softness due to coercivity decrease of about 30%. Annealed sample have substantial reduced power loses in mean frequency domain and still poses the high remanence ratio (about 0.8) making this alloy perspective for signal processing devices.

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Садржај: *У раду је испитан утицај температуре и фреквенције на пермеабилност, коерцитивну силу и 'магнетне' губитке аморфне траке $Fe_{81}B_{13}Si_4C_2$. Мерења магнетне пермеабилности, која су спроведена у изотермским и неизотермским условима, потврдила су да се процес структурне релаксације одиграва на 663 К. Овај процес се одвија двостепено: први ступањ је кинетички, са енергијом активације E_{a1}=52.02 kJ/mol, а други ступањ је дифузиони, са енергијом активације*

E_{a2}=106.9kJ/mol. Термичким одгревањем на 663 K постигнуто је побољшање коерцитивне силе од око 30 %, као и значајно смањење магнетних губитака. На основу анализе фреквентних карактеристика магнетне пермеабилности дошло се до закључка да се испитивана легура $Fe_{81}B_{13}Si_4C_2$ може користити у направама за процесирање сигнала у домену средњих фреквенција.

Кључне речи: *Аморфна легура; структурна релаксација; одгревање; побољшање магнетномеких карактеристика*