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Changes of properties during the annealing $Fe_{28}Co_{50}Si_9B_{13}$ and $Fe_{28}Ni_{50}Si_9B_{13}$ metallic glasses

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The crystallization of $Fe_{28}Co_{50}Si_9B_{13}$ and $Fe_{28}Ni_{50}Si_9B_{13}$ metallic glasses was stimulated by isochronal annealing at the temperature of 573–823 K, and was investigated by measuring the electrical resistivity, Hall effect and X-ray diffraction. The substitution of Co by Ni has proved the differences in the initial and final parameters, as well as in the kinetics of crystallization. The creation of crystalline phases from the amorphous matrix was related to the abrupt decrease in the electrical and Hall resistivities, and in the spontaneous Hall coefficient.

1. Introduction

Amorphous alloys have different parameters, compared with alloys of the same composition in the crystalline state. Moreover, some properties such as elasticity (Young modulus), volume, magnetic coercion, electrical resistivity, Curie temperature and specific heat are particularly sensitive to the structural changes [1], [2]. The amorphous alloys, due to the transition to the crystalline state, decrease distinctly their electrical resistivity [3]–[5]. Therefore, metallic glasses are often applied to the technique of high frequencies because they decrease losses caused by eddy currents. Recently, more and more often the metallic glasses based on Fe or Co have been applied to the production of transformers [6].

In the production of metallic glasses, the alloy atoms in transition from the liquid to the solid state do not take the equilibrium position and do not reach the minimum of the energy. As a result, the amorphous state is a metastable one and it evolves through changes in the short and intermediate range order to the polycrystalline state [7]. The initiation process and the crystallization kinetics depend on many factors like the number of "frozen" crystallization centres, stress, state of the surface, diffusion, composition of the alloy, as well as conditions stimulating the structural changes. For the determined composition, the above parameters are usually different and the determined crystallization temperature is not typical of the given alloy composition.

The aim of the paper was to investigate the crystallization of $Fe_{28}Co_{50}Si_9B_{13}$ and $Fe_{28}Ni_{50}Si_9B_{13}$ metallic glasses and to determine the influence of the alloy composition (*i.e.*, the substitution of Co by Ni) on the kinetics and the crystallization temperature, as well as on the initial and final parameters of alloys. These investigations were carried out by the methods of Hall effect, electrical resistivity, and X-ray diffraction.

2. Experimental

The $Fe_{28}Co_{50}Si_9B_{13}$ and $Fe_{28}Ni_{50}Si_9B_{13}$ metallic glasses were prepared by the melt spinning technique (Institute of Materials Engineering of Warsaw Technical University, Poland). The electrical and Hall resistivities and X-ray diffraction measurements were performed for the as-received as well as isochronal (for 4 h) annealed samples at the temperatures 573, 673, 723, 773 and 823 K in an argon atmosphere. The Hall voltage was measured by a constant current method in the field up to 3.26 T. The electrical resistivity was also measured within d.c. regime. The samples for the measurements of the Hall and electrical resistivities were prepared by selective etching using photolitography. The X-ray studies were performed using a DRON-2.0 diffractometer with a horizontal goniometer of GUR-5 type. The X-ray tube had a molybdenium target ($\lambda_{K\alpha} = 0.71069\times10^{-10}$ m) and a graphite monochromator in the primary beam.

3. Results and discussion

The measurements of electrical resistivity changes during the transition from the as-received amorphous state to the crystalline one are presented in Fig. 1. They are presented as a relative change of the electrical resistivity $\Delta\rho/\rho_0$ (*i.e.*, related to the resistivity of the as-received samples ρ_0) *vs.* the isochronal annealing temperature. For $Fe_{28}Co_{50}Si_9B_{13}$ and $Fe_{28}Ni_{50}Si_9B_{13}$ the values of ρ_0 are 1.74±0.03 μΩm and 1.33±0.02 μΩm, respectively. The substitution of Co by Ni decreases ρ_0 and consequently $\Delta\rho/\rho_0$.

Figures 2 and 3 present results of the Hall resistivity ρ_H *vs.* the external magnetic induction B_0 for samples in the as-received state, as well as samples annealed isochronally at different temperatures. With the increase of the annealing temperature the curves fall and a distinct decrease occurs after the annealing at temperatures of 723 K and 773 K. The crystallized phases do not change the final macroscopic ferromagnetic state of the substances. All the curves in Figs. 2 and 3 are typical of the ferromagnetic materials and are described by an equation [8]

$$\rho_H = R_o B_0 + R_s M(B_0) \tag{1}$$

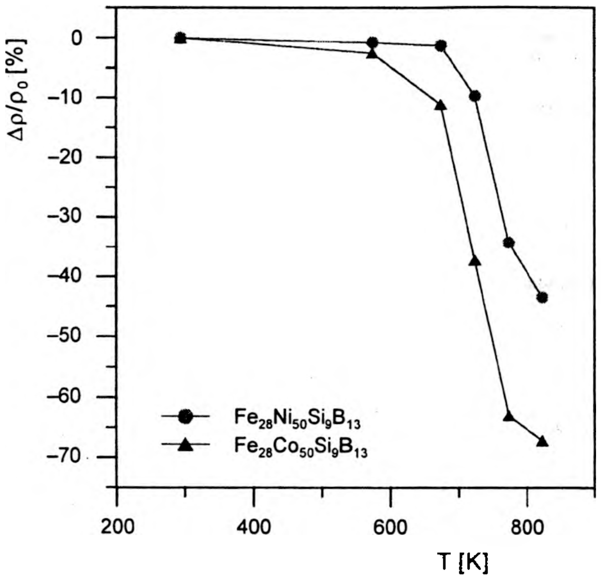

Fig. 1. Relative electrical resistivity $\Delta\rho/\rho_0$ as a funtion of annealing temperature T for $Fe_{28}Co_{50}Si_9B_{13}$ and $Fe_{28}Ni_{50}Si_9B_{13}$ alloys.

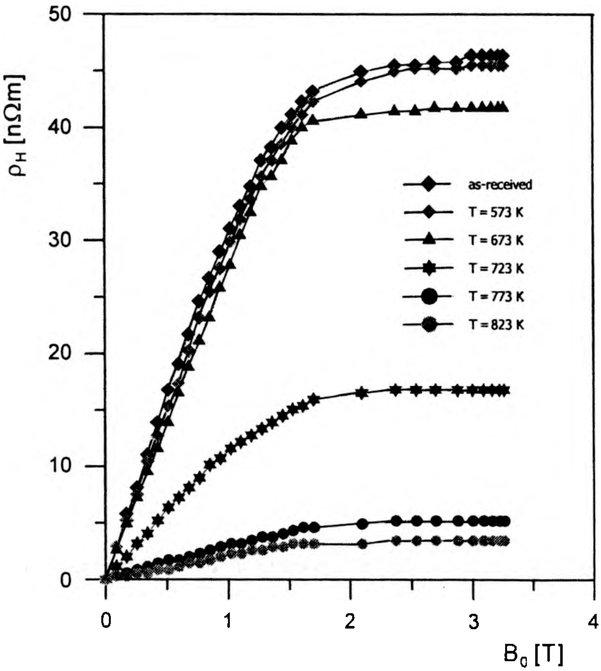

Fig. 2. Hall resistivity ρ_H *vs.* applied magnetic induction B_0 for annealed $Fe_{28}Co_{50}Si_9B_{13}$ alloy.

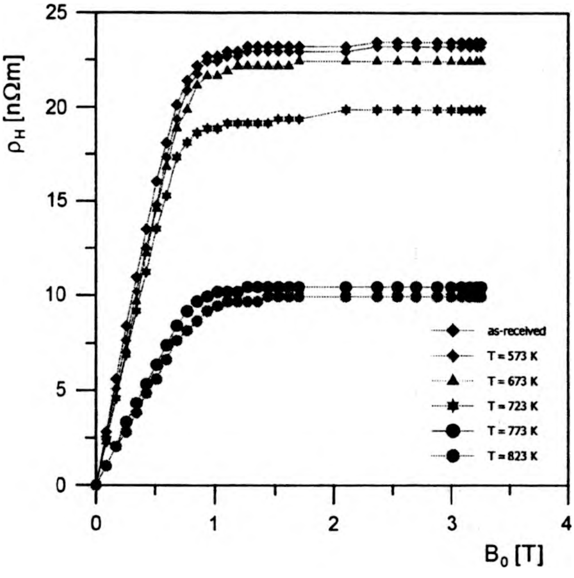

Fig. 3. Hall resistivity ρ_H *vs*. applied magnetic induction B_0 for annealed $Fe_{28}Ni_{50}Si_9B_{13}$ alloy.

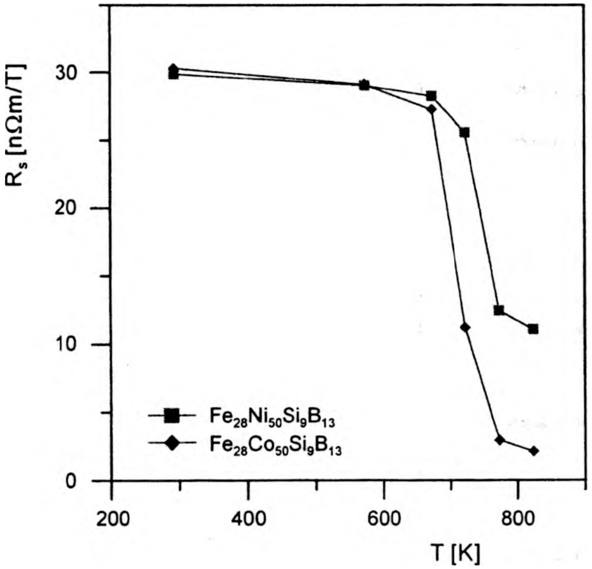

Fig. 4. Spontaneous Hall coefficient R_s as a function of annealing temperature T for $Fe_{28}Co_{50}Si_9B_{13}$ and $Fe_{28}Ni_{50}Si_9B_{13}$ alloys.

where R_o and R_s are the ordinary and spontaneous Hall coefficients, respectively, and $M(B_0)$ is magnetization of the sample. The Hall resistivity $\rho_H = E_y/j_x$ is the transverse field E_y, called the Hall field, per unit longitudinal current density j_x. The field E_y is created when the current with density j_x flows through a sample placed in the external magnetic field $\mathbf{B}_0(0,0,B_z)$ normal to it. The first component of Eq. (1) is connected with the action of the Lorentz force on the current carriers. The second component represents the initial part of the curves. It is connected with a ferromagnetic state and derives from different mechanisms: skew scattering, side jump mechanism, spin dependent scattering and the transition from the low-field regime to the high-field regime. Figure 4 shows a graph of the dependence of R_s on the annealing temperature T. The abrupt decrease of the R_s value occurs after annealing at the temperatures 723 K and 773 K. The temperature of the sharp drop of R_s values with the constant rise in sample temperature ($\Delta T/\Delta\tau = \text{const}$) is known as Curie temperature [9]. However, the abrupt R_s decrease found in this investigation can be explained by structural changes, leading to the decrease in the asymmetrical scattering on the magnetic moments of the substance. The result of these changes are the phase changes of the first type, because the crystallized phases do not change the final macroscopic ferromagnetic state of the substances (Figs. 2 and 3).

Table. Phases indentified after crystallization of $Fe_{28}Co_{50}Si_9B_{13}$ and $Fe_{28}Ni_{50}Si_9B_{13}$ metallic glasses.

	$Fe_{28}Co_{50}Si_9B_{13}$					$Fe_{28}Ni_{50}Si_9B_{13}$			
Phase	a [Å]	b [Å]	c [Å]	V [Å^3]	Phase	a [Å]	b [Å]	c [Å]	V [Å^3]
α-Fe(Co)	2.845	—	—	23.05	Ni(Fe)	3.567	—	—	46.15
$(Fe, Co)_3B$	5.215	6.673	4.37	152.17	$(Ni, Fe)_3B$	5.26	6.64	4.42	154.6
$(Fe, Co)_2B$	5.116	—	4.191	109.72					

To identify the quality and abundance of the created crystalline phases, the X-ray diffraction was carried out for the samples undergoing the same thermal treatment. The X-ray patterns confirmed the results of the electrical and Hall resistivities investigation, *i.e.*, the annealing at the temperatures of 723 and 773 K led to the phase changes of the first type. The results of the phase quality analysis performed for the samples annealed at the temperature of 823 K are shown in the Table [10]. At the temperature of 673 K the alloys underwent isothermal annealing at a different time of $\tau \leqslant 2\times10^4$ s and it has been stated that for $Fe_{28}Co_{50}Si_9B_{13}$ alloy the creation of the α-Co(Fe) phase begins after the annealing at time $\tau = 2\times10^4$ s; for $Fe_{28}Ni_{50}Si_9B_{13}$ the phase Ni(Fe) is created at time $\tau = 10^4$ s and finally in both alloys there is 80% of these phases.

4. Conclusions

The crystallization of $Fe_{28}Co_{50}Si_9B_{13}$ and $Fe_{28}Ni_{50}Si_9B_{13}$ metallic glasses is a two stage process. In the first one (723 K) a creation of α-Co(Fe) and Ni(Fe) phases appears, and in the second one (773 K) a formation of $(Fe,Co)_2B$, $(Fe,Co)_3B$ and $(Ni,Fe)_3B$, phases takes place in the alloys containing Co and Ni.

Creation of the crystalline phase out of the amorphous matrix is related to the abrupt decrease in the values of the electrical and Hall resistivities and the spontaneous Hall coefficient.

The substitution of Co by Ni in the alloys decreases the electrical and Hall resistivities in the as-received state and causes smaller changes of their values as a result of the annealing.

The amorphous state of the alloy containing Ni is less stable.

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