Optimisation the magnetic properties of the (Fe₁₋ₓCoₓ)₇₃.₅Cu₁Nb₃Si₁₃.₅B₉ (X=10;30;40) alloys

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ABSTRACT

Purpose: In this paper results of experience of influence a structure (amorphous and amorphous after structural relaxation) on the magnetic properties (the initial magnetic permeability μ_p, the magnetic permeability μ in function magnetic field H, the coercive field Hc, the remanence Br and the saturation induction Bs) on $(Fe_{1-X}Co_X)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (X=10;30;40) alloys have been presented.

Design/methodology/approach: The material was obtained by the method of rapid cooling from liquid phase. The measurements of structure were made on the magnetic balance, the X-ray diffraction and the mössbauer measurements. The measurements of the magnetic properties were made on the Maxwell-Wien bridge, the fluxometr and the VSM - Vibrating Sample Magnetometer.

Findings: The results allowed defining that are significant dependence between the magnetic properties and the structure of the material.

Research limitations/implications: Due to the high influence of the cobalt on the magnetic properties of the material further research should be undertaken.

Practical implications: The measurements of allows give information to the industry how affect on the magnetic properties of alloys by adequate the heat treatment.

Originality/value: The Finemet is very attractive due to his excellent the soft magnetic properties. The problem of his application in higher temperature of application and improvement his magnetic properties have been presented.

Keywords: Amorphous materials, Finemet type alloys, Magnetic properties, Mössbauer analysis

1. Introduction

The soft magnetic materials have in their the chemical constitution the iron and other elements, which decide about the magnetic properties and the adequate kinetics of the crystallization. From reason that is many the chemical composition there is not general classification of names and occurring names accrue from producers. Nowadays use names are FINEMET, NANOPERM and HITPERM [1-5] and different variants dependently of the chemical constitution.

The modification of the chemical composition often makes to improve the magnetic properties Alloys obtain by using by spinning technology have amorphous and nanocrystalline structure. Often the amorphous ribbons have submitted heating process to aim the nanocrystalline structure. The structure is depended on the chemical composition and parameters of the technological processes [5-9].

The Finemet is an alloy with very good magnetic properties. Its great scientific interest is not only because of its suitability for many technological applications. It constitutes a unique object for fundamental studies of magnetism. Partial substitution of the iron by the cobalt in nanocrystalline Finemet–type alloys allows improving very good soft magnetic properties in elevated temperatures [10-15].

In this paper the results of experience of influence a structure on the magnetic properties of $(Fe_{1-X}Co_X)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (X=10;30;40) alloys have been presented.

2. Experimental

The researches include $(Fe_{1-X}Co_X)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (X=10; 30; 40) alloys in state „as quenched" obtained in the form of strips (thickness of about 25 μm) by the method of rapid cooling from liquid phase.

The strips of researched materials in state „as quenched" were isothermally heat treated (denoted as a heat treatment temperature T_a) in range 473-923 K with the step 50 K, for 1 h. In a range of 773-798 K with the step 25 K when was expected the best magnetic permeability μ. The samples were heat treated in an electrical chamber furnace Thermolyne Model No.F 6020C-60 with an argon atmosphere.

A structure of the researched $(Fe_{1-X}Co_X)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (X=10; 30; 40) alloys by using the X-ray diffractometer XRD 7 of firm Seifert-FPM and the mössbauer measurements of firm POLON was determinate. To analyse a spectrum of the mössbauer measurements was using program Normos, which allow decomposing spectrum of the mössbauer on components spectrum and appointing distribution of hyperfine fields.

A measurement volume of magnetisation in function of the heat treatment temperature was made to define the temperature of the primary crystallisation and the Curie temperature. The measurements using the magnetic balance. The measurements used to assign a magnetisation in a function of intensity magnetic field were designated in a protect atmosphere - helium, with rate of the worm up 10 K/min.

The initial magnetic permeability $μ_p$ defined using the Maxwell-Wien bridge - working within a weak magnetic field (about 0.5 A/m) of frequency about 1 kHz. Electric circuit of bridge was feeded by AC from generator of acoustic oscillator. A null indicator is selectivity analyser of harmonic voltage. A microvoltmeter with high internal resistance is parallel connected with resistance R_1. These allows observing a decrease of circuit and attuning a magnetic field.

The magnetic permeability μ in function of magnetic field H was measured by the fluxometr. The material was put to a field coil. The main element of the system is the fluxometr (F) which allow measuring of changes magnetic flux in the sample of researched material.

The other magnetic properties (the coercive field H_c, the remanence B_r and the saturation induction B_s) were measured using apparatus the VSM – Vibrating Sample Magnetometer. The researched sample was impelled in vibrations by piezoelectric transducer. This apparatus was in coils which were between a field of electromagnets. The Hall probe used to measurements records induction of the magnetic field, in which the sample was during induced the magnetic moment. Voltage in the coils was proportional to the magnetic moment investigated sample and appeared from magnetic flux.

3. Results and discussion

The research using the X-ray diffraction allowed defining quality changes in the material structure caused by the heat treatment $(Fe_{63.5}Co_{10}Cu_1Nb_3Si_{13.5}B_9$, $Fe_{43.5}Co_{30}Cu_1Nb_3Si_{13.5}B_9$, $Fe_{33.5}Co_{40}Cu_1Nb_3Si_{13.5}B_9)$ alloys originally in the state „as quenched".

The X-ray diffraction of the alloy defined that the strip of material obtained by the method of rapid cooling from liquid phase has the amorphous structure. It was observed one characteristic widened diffraction line in a place where is observed a diffraction line from plane (110) on the diffraction pattern, figure 1.
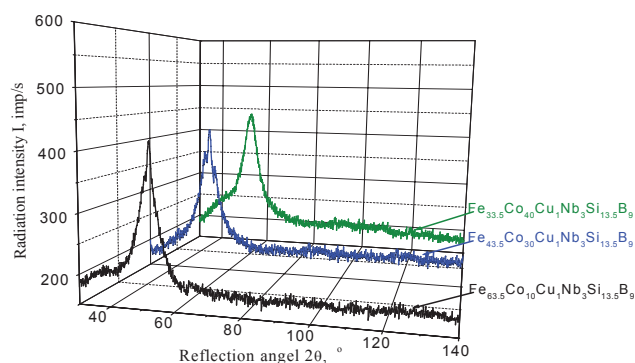

Fig. 1. The X-ray diffraction pattern for the $(Fe_{1-X}Co_X)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (X=10;30;40) alloys in state "as quenched"

For the material after the heat treatment in range of the temperature 473-798 K the diffraction pattern doesn't demonstrated the crystal structure. The diffraction pattern also doesn't show changes in intensity in the diffraction lines in a place when is observed a diffraction line from plane (110) α-Fe, figure 2.
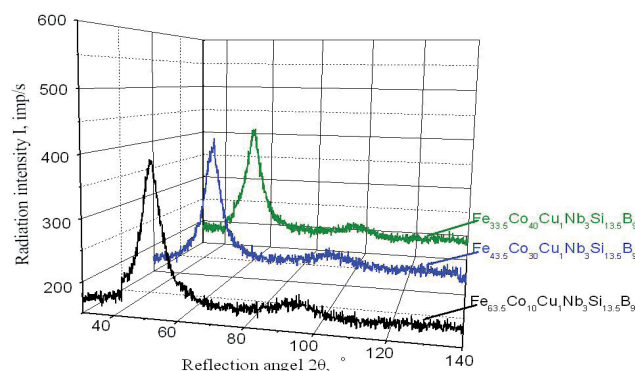

Fig. 2. The X-ray diffraction pattern for the $(Fe_{1-X}Co_X)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (X=10;30;40) alloys after heat treatment at temperature 798 K

The mössbauer research allowed to identify a phase which accrue during the heat treatment the amorphous $(Fe_{1-X}Co_X)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (x=10; 30; 40) alloys.

A shape of spectrum and a hyperfine field obtain for researched alloys in state „as quenched" are typical the spectrum of ferromagnetic amorphous alloys. A variety a surroundings atom of the iron in the amorphous structure causes broadening and a superimposing six magnetic lines of the hyperfine structure in the spectrum which characterizes wide a distribution of the hyperfine magnetic field H. The hyperfine field allocated for the amorphous alloys is displayed as a wide peaks replay the amorphous phase. An average value the hyperfine field B for the amorphous phase are presented in table 1, figure 3.

Table 1.
The value of average hyperfine field - $(B_{eff})_{amorf}$, T; the standard deviation for the spectrum of effective induction the hyperfine field - D_{amorf}, T; proportion intensity 2 and 5 to 3 and 4 respective sextets referred from surroundings atoms of iron - $A_{2,5}$, the relative intensity a part of spectrum response a amorphous phase - Int_{amorf}, researched alloys in state „as quenched"

Type of the alloys	Alloy in state „as quenched"		
	$Fe_{63.5}Co_{10}Cu_1Nb_3Si_{13.5}B_9$	$Fe_{43.5}Co_{30}Cu_1Nb_3Si_{13.5}B_9$	$Fe_{33.5}Co_{40}Cu_1Nb_3Si_{13.5}B_9$
$(B_{eff})_{amorf}$	20.60	21.03	20.57
D_{amorf}	5.03	4.88	5.00
$A_{2,5}$	2.4	2.4	2.4
Int_{amorf}	1	1	1

Table 2.
The value of average hyperfine field - $(B_{eff})_{amorf}$, T; the standard deviation for the spectrum of effective induction the hyperfine field - D_{amorf}, T; proportion intensity lines 2 and 5 to 3 and 4 respective sextets referred from surroundings atoms of iron - $A_{2,5}$, the relative intensity a part of spectrum response a amorphous phase - Int_{amorf}, researched alloys after heat treatment at temperature 798 K

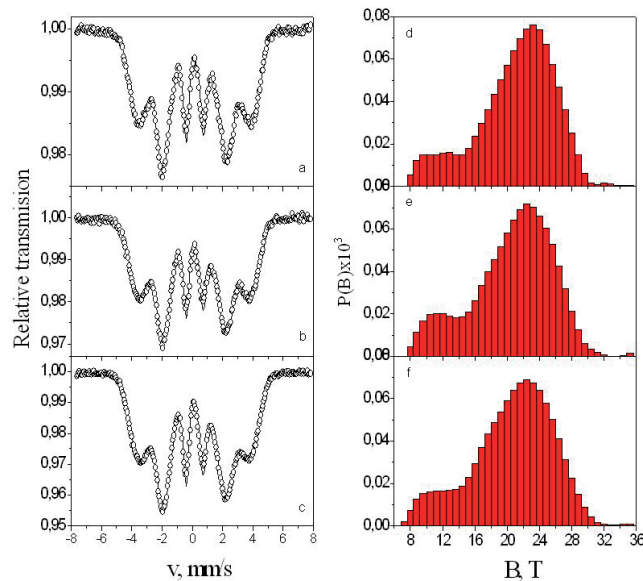| Type of the alloys	Alloy in state after heat treatment at temperature 798 K		
	$Fe_{63.5}Co_{10}Cu_1Nb_3Si_{13.5}B_9$	$Fe_{43.5}Co_{30}Cu_1Nb_3Si_{13.5}B_9$	$Fe_{33.5}Co_{40}Cu_1Nb_3Si_{13.5}B_9$
$(B_{eff})_{amorf}$	21.12	22.00	21.49
D_{amorf}	4.92	4.71	4.85
$A_{2,5}$	3.4	2.7	2.7
Int_{amorf}	1	1	1

Fig. 3. The mössbauer spectrum (a, b, c) and respond them distribution of the hyperfine field on iron nucleus ^{57}Fe (d, e, f) obtained for the $(Fe_{1-x}Co_x)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (X=10; 30; 40) alloys in state „as quenched", where: „a" and „d" for the $Fe_{63.5}Co_{10}Cu_1Nb_3Si_{13.5}B_9$ alloy, „b" and „e" for the $Fe_{43.5}Co_{30}Cu_1Nb_3Si_{13.5}B_9$ alloy, „c" and „f" for the $Fe_{33.5}Co_{40}Cu_1Nb_3Si_{13.5}B_9$ alloy

The shape of the mössbauer spectrum researched alloys after the heat treatment at the temperature 798 K - below the temperature T_{x1}, shows the changes of average value of the hyperfine B amorphous phase in proportion to the spectrum samples in state "as quenched" are noted. For each of researched alloys the increase of the value $(B_{eff})_{amorf}$ is noted. It is connected with increase an order in the surroundings atom of iron in an amorphous structure. Additionally changes in value of the $(B_{eff})_{amorf}$ can be caused by decay a free volume in process of the structural relaxation at the same time decreasing a internal stresses by the relaxation them, table 2, figure 4.
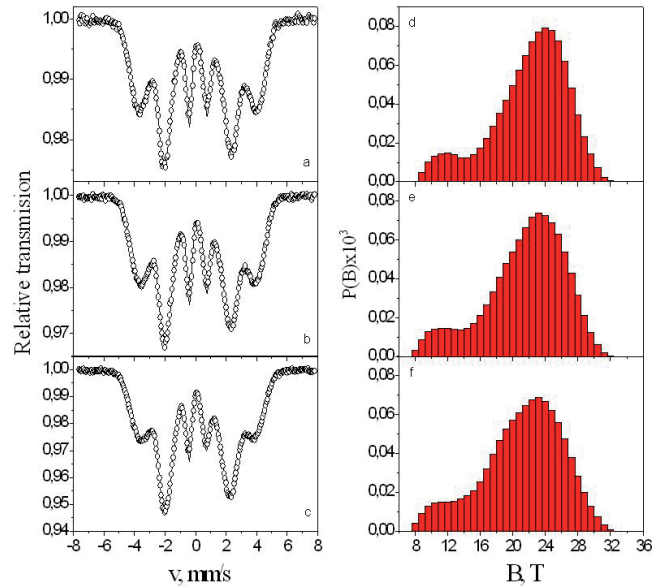

Fig. 4. The mössbauer spectrum (a, b, c) and respond them distribution of the hyperfine field on iron nucleus ^{57}Fe (d, e, f) obtained for the $(Fe_{1-x}Co_x)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (X=10; 30; 40) alloys heat treatment at the temperature 798 K, where: „a" and „d" for the $Fe_{63.5}Co_{10}Cu_1Nb_3Si_{13.5}B_9$ alloy, „b" and „e" for the $Fe_{43.5}Co_{30}Cu_1Nb_3Si_{13.5}B_9$ alloy, „c" and „f" for the $Fe_{33.5}Co_{40}Cu_1Nb_3Si_{13.5}B_9$ alloy

Results of measurements of the magnetisation ratio in function of the heat treatment temperature the $(Fe_{1-X}Co_X)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (X=10;30;40) alloys allowed determine the temperature of the primary crystallisation T_x and the Curie temperature T_c^{am}, figure 5, 6.

On the beginning of the heat treatment the samples in range of temperatures 300-700 K the magnetisation of amorphous phase decrease. It is connected with transformation phase ferromagnetic - paramagnetic, getting near null, figure 5. This point respond to minimal value of $\frac{dM}{dT}$ (denoted as T_c^{am}) and also is noted as the Curie temperature, figure 6. The Curie temperature T_c^{am} for researched alloys is presented in table 3.

A low magnetisation of the material in paramagnetic phase is until moment as start the crystallisation of the amorphous phase. Formation of the new ferromagnetic phase in an amorphous matrix causes increasing of the value of magnetisation in higher temperature of the heat treatment, table 3.
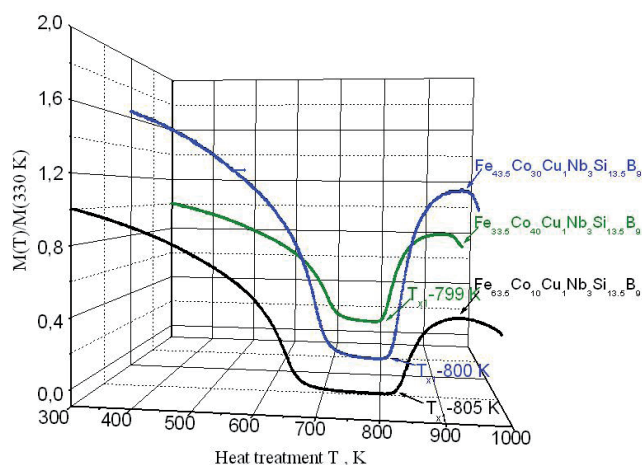
Fig. 5. Normalised curves of the magnetisation determined in a function of the heat treatment temperature for the $(Fe_{1-X}Co_X)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (X=10; 30; 40) alloys
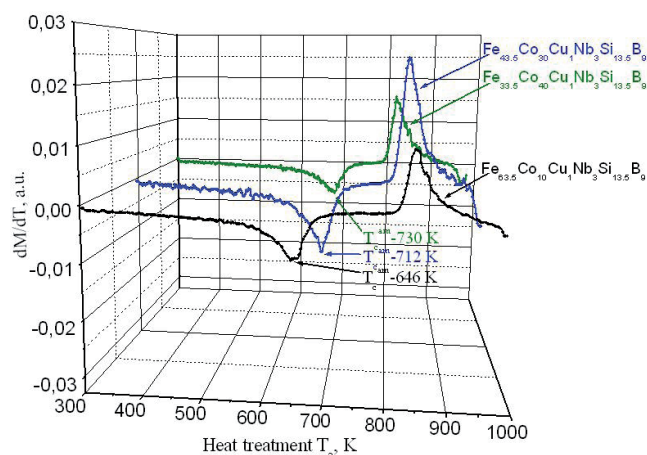

Fig. 6. Dependence dM/dT for normalised curves of magnetisation determined in function of the heat treatment temperature for the $(Fe_{1-X}Co_X)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (X=10 ;30; 40) alloys

Table 3.
The crystallisation temperature T_{x1} and the Curie temperature T_c^{am} of $(Fe_{1-X}Co_X)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (X=10; 30; 40) alloys with rate of the heat treatment 10 K/min

Type of the alloy	Crystallisation temperature T_{x1}, K	Curie temperature T_c^{am}, K
$Fe_{63.5}Co_{10}Cu_1Nb_3Si_{13.5}B_9$	805	646
$Fe_{43.5}Co_{30}Cu_1Nb_3Si_{13.5}B_9$	800	712
$Fe_{33.5}Co_{40}Cu_1Nb_3Si_{13.5}B_9$	799	730

After the heat treatment at the temperatures 473-923 K changes of the magnetic properties were observed in two steps, figure 7. Firstly the magnetic properties increase in to their value in state „as quenched" and later they decrease. After the heat

treatment in range of the temperature 473-723 K the initial magnetic permeability μ_p slowly increases but at the temperature 723 K is observed strong increases this property. Researched alloys have the best initial magnetic permeability at the temperature 798 K, which is noted as an optimal temperature of the heat treatment - T_{op}, table 4. The ribbons with increase the value of cobalt in their chemical constitution have worse the initial magnetic permeability.
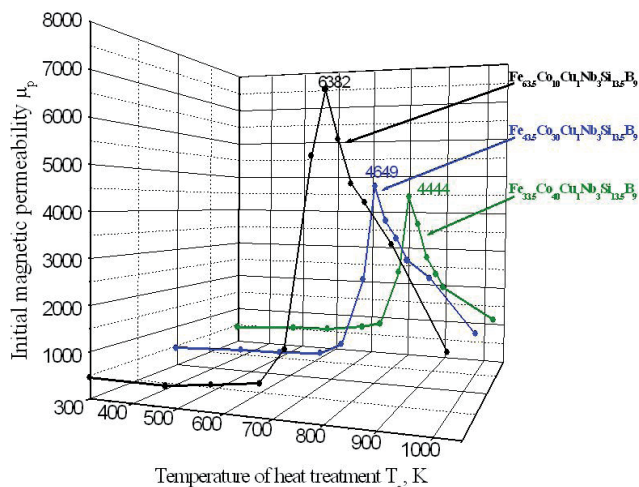

Fig. 7. The initial magnetic permeability μ_p the $(Fe_{1-X}Co_X)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (X=10; 30; 40) alloys in function of the heat treatment

Table 4.
The value of maximum the initial magnetic permeability μ_p the $(Fe_{1-x}Co_x)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (X=10; 30; 40) alloys

Type of the alloys	The value of maximum the initial magnetic permeability μ_p
$Fe_{63.5}Co_{10}Cu_1Nb_3Si_{13.5}B_9$	6382
$Fe_{43.5}Co_{30}Cu_1Nb_3Si_{13.5}B_9$	4649
$Fe_{33.5}Co_{40}Cu_1Nb_3Si_{13.5}B_9$	4444

Research of the magnetic permeability μ in function of magnetic field H the $(Fe_{1-x}Co_x)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (x=10; 30; 40) alloys allowed to designate the maximum of magnetic permeability in inflicted magnetic field H. For the ribbon in state „as quenched" and after the heat treatment in the temperature 798 K, which responds the best initial magnetic permeability μ_p, figure 8.

The ribbon of the $Fe_{63.5}Co_{10}Cu_1Nb_3Si_{13.5}B_9$ alloy in state „as quenched" indicates a low value of the magnetic permeability μ. Gradual increase the intensity of the magnetic field during the research causes the increase magnetic permeability, which maximum value for the $Fe_{63.5}Co_{10}Cu_1Nb_3Si_{13.5}B_9$ alloy is observed in the magnetic field - 33.33 A/m and the value is 1517, table 5. But in the same magnetic field - 33.33 A/m for the $Fe_{43.5}Co_{30}Cu_1Nb_3Si_{13.5}B_9$ alloy characterize the maximum magnetic permeability, which is 1349. For the

$Fe_{33.5}Co_{40}Cu_1Nb_3Si_{13.5}B_9$ alloy is observed analogical apparition causes the decrease magnetic permeability to the value 1316, table 5. The increasing magnetic field cause decreasing the value of the magnetic field for each of research alloys, figure 8.

The ribbons after the heat treatment in the temperature 798 K, which characterize the best initial magnetic permeability, have the better magnetic permeability μ in their value in state "as quenched", figure 8. For the magnetic field - 26.67 A/m the ribbon from the $Fe_{63.5}Co_{10}Cu_1Nb_3Si_{13.5}B_9$ alloy has maximum magnetic permeability - 7348, table 6. For other research alloys in the magnetic field - 33.33 A/m the magnetic permeability is for alloys: $Fe_{43.5}Co_{30}Cu_1Nb_3Si_{13.5}B_9$ - 6582 and $Fe_{33.5}Co_{40}Cu_1Nb_3Si_{13.5}B_9$ - 6385, table 6. The magnetic field H inflicted more than 33.33 A/m causes gradually decrease the value of the magnetic permeability μ.
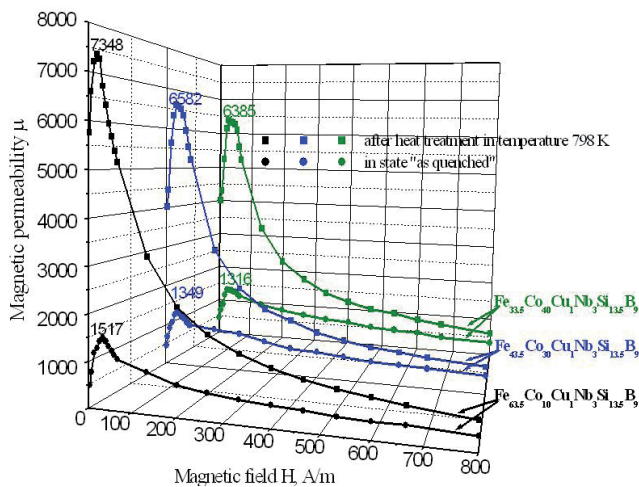

Fig. 8. The magnetic permeability μ the $(Fe_{1-x}Co_x)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (X=10; 30; 40) alloys in state "as quenched" and after the heat treatment in temperature 798 K in function of the magnetic field H

Table 5.
The value of the maximum magnetic permeability μ the $(Fe_{1-x}Co_x)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (X=10; 30; 40) alloys in state "as quenched" in function the magnetic field H

Type of the alloys	The maximum magnetic permeability μ	The value of the magnetic field, A/m
$Fe_{63.5}Co_{10}Cu_1Nb_3Si_{13.5}B_9$	1517	33.33
$Fe_{43.5}Co_{30}Cu_1Nb_3Si_{13.5}B_9$	1349	33.33
$Fe_{33.5}Co_{40}Cu_1Nb_3Si_{13.5}B_9$	1316	33.33

The researches of the magnetic properties of the $(Fe_{1-x}Co_x)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (X= 10; 30; 40) alloys: the coercive field H_C, the remanence B_r and the saturation induction B_S in state "as quenched" and after the heat treatment at temperature, where is effect of the structural relaxation, were done, table 7.

Table 6.
The value of the maximum magnetic permeability μ the $(Fe_{1-x}Co_x)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (x=10; 30; 40) alloys with the structure amorphous after the structural relaxation at temperature 798 K in function the magnetic field H

Type of the alloys	The maximum magnetic permeability μ	The value of the magnetic field, A/m
$Fe_{63.5}Co_{10}Cu_1Nb_3Si_{13.5}B_9$	7348	26.67
$Fe_{43.5}Co_{30}Cu_1Nb_3Si_{13.5}B_9$	6582	33.33
$Fe_{33.5}Co_{40}Cu_1Nb_3Si_{13.5}B_9$	6385	33.33

The ribbons of the $Fe_{63.5}Co_{10}Cu_1Nb_3Si_{13.5}B_9$ alloy in state "as quenched" has following magnetic properties: the coercive field H_C - 23.221 A/m, the remanence B_r - 0.111 T and the saturation induction B_S - 0.987 T. Increasing the cobalt in alloys cause decreasing the magnetic properties and for the $Fe_{43.5}Co_{30}Cu_1Nb_3Si_{13.5}B_9$ alloy: the coercive field H_C - 25.764 A/m, the remanence B_r - 0.097 T and the saturation induction B_S - 0.979 T, and for the $Fe_{33.5}Co_{40}Cu_1Nb_3Si_{13.5}B_9$ alloy: the coercive field H_C - 27.157 A/m, the remanence B_r - 0.093 T and the saturation induction B_S - 0.962 T, table 7. Researched ribbons in state "as quenched" characterize difference in the magnetic properties between the examined alloys.

Table 7.
The magnetic properties of the $(Fe_{1-x}Co_x)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (X=10; 30; 40) alloys

Type of alloys	Temperature of the heat treatment, K			
	"as quenched"	673	773	798
Coercive field H_C, A/m				
$Fe_{63.5}Co_{10}Cu_1Nb_3Si_{13.5}B_9$	23.221	8.644	2.497	2.462
$Fe_{43.5}Co_{30}Cu_1Nb_3Si_{13.5}B_9$	25.764	11.117	3.359	3.810
$Fe_{33.5}Co_{40}Cu_1Nb_3Si_{13.5}B_9$	27.157	12.925	4.852	5.230
Remanence B_r, T				
$Fe_{63.5}Co_{10}Cu_1Nb_3Si_{13.5}B_9$	0.111	0.123	0.015	0.037
$Fe_{43.5}Co_{30}Cu_1Nb_3Si_{13.5}B_9$	0.097	0.088	0.014	0.059
$Fe_{33.5}Co_{40}Cu_1Nb_3Si_{13.5}B_9$	0.093	0.069	0.011	0.061
Saturation induction B_S, T				
$Fe_{63.5}Co_{10}Cu_1Nb_3Si_{13.5}B_9$	0.987	1.187	1.200	1.210
$Fe_{43.5}Co_{30}Cu_1Nb_3Si_{13.5}B_9$	0.979	1.145	1.156	1.170
$Fe_{33.5}Co_{40}Cu_1Nb_3Si_{13.5}B_9$	0.962	1.136	1.142	1.157

The researches of the magnetic properties of the alloys after the heat treatment at temperature from 673 K to 798 K indicate improve the magnetic properties in to state "as quenched". The $Fe_{63.5}Co_{10}Cu_1Nb_3Si_{13.5}B_9$ alloy at the temperature 798 K, which characterizes the best initial magnetic permeability, has the best

magnetic the coercive field from all temperatures range of heat treatment. The coercive field is H_C - 2.462 A/m, remanence B_r - 0.037 T and saturation induction B_S - 1.210 T, table 7. But for other alloys: $Fe_{43.5}Co_{30}Cu_1Nb_3Si_{13.5}B_9$, $Fe_{33.5}Co_{40}Cu_1Nb_3Si_{13.5}B_9$ the best magnetic properties are at the temperature 773 K. This is temperature about 25 K low than for the $Fe_{63.5}Co_{10}Cu_1Nb_3Si_{13.5}B_9$ alloy. For the $Fe_{43.5}Co_{30}Cu_1Nb_3Si_{13.5}B_9$ alloy the value of the coercive field is H_C - 3.359 A/m also is lower remanence B_r - 0.014 T and better the saturation induction B_S - 1.156 T. For the $Fe_{33.5}Co_{40}Cu_1Nb_3Si_{13.5}B_9$ alloy the value of the coercive field - H_C - 4.852 A/m, remanence B_r - 0.011 T and better the saturation induction B_S - 1.142 T, table 7. Difference between the values of the optimal temperature of heat treatment is caused by the temperature of crystallization. The alloys with more the cobalt in the chemical constitution have the temperature crystallization lower than the $Fe_{63.5}Co_{10}Cu_1Nb_3Si_{13.5}B_9$ alloy.

4. Conclusions

- The results of the research the $(Fe_{1-X}Co_X)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (X=10; 30; 40) alloys indicate a big dependence between the magnetic properties and the structure of alloys,
- The optimal magnetic properties of the alloys were obtained for the amorphous structure after the structural relaxation in the temperature T_{op} - 798 K ,
- The best magnetic properties are accomplished for the $Fe_{63.5}Co_{10}Cu_1Nb_3Si_{13.5}B_9$ alloy.

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