

## Structure and magnetic properties of $\text{Fe}_{63.5}\text{Co}_{10}\text{Cu}_1\text{Nb}_3\text{Si}_{13.5}\text{B}_9$ alloy

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Received 15.03.2006; accepted in revised form 30.04.2006

### Materials

#### ABSTRACT

**Purpose:** In this paper results of experience of influence a structure (amorphous, amorphous after structural relaxation and nanocrystal) and influence a corrosion medium, which was 1 M solution NaCl, on magnetic properties (initial magnetic permeability  $\mu_p$ , relative magnetic permeability  $\mu_w$ , coercive field  $H_c$ , remanence  $B_r$  and saturation magnetisation  $B_s$ ) on  $\text{Fe}_{63.5}\text{Co}_{10}\text{Cu}_1\text{Nb}_3\text{Si}_{13.5}\text{B}_9$  alloy 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. The measurements of magnetic properties were made on the Maxwell-Wien bridge, the fluxometer and the VSM – Vibrating Sample Magnetometer.

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

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

**Practical implications:** The measurements allow giving information to the industry how decrease the magnetic properties of alloy after influence the corrosion medium.

**Originality/value:** The Finemet is very attractive due to his excellent soft magnetic properties. The problem of the corrosion has been presented and her influence on the magnetic properties.

**Keywords:** Amorphous materials; X-ray phase analysis; Finemet type alloys; Corrosion

### 1. Introduction

Amorphous and nanocrystalline alloys contain iron and cobalt are known as a soft magnetic materials. These alloys very often are made using technology consists two operations. In first operation amorphous ribbon was made by vitrification metallic fluid on whirling rollers. In second operation nanocrystalline structure is created after primary crystallisation using appropriate parameters [1-3]. A possibility of creating amorphous and nanocrystalline structure is connected with appropriate chemical composition. Very good magnetic properties amorphous and nanocrystalline alloys are acquired due to a lot of modification chemical composition. This allowed wresting some kind of material group, as a: FINEMET, NANOPERM and HITPERM

[4-9]. Alloy called FINEMET has attracted great scientific interest, not only because of its suitability for many technological applications but as it constitutes a unique object for fundamental studies of magnetism. Partial substitution of Fe by Co in nanocrystalline Finemet-type alloys allows improving very good soft magnetic properties in elevated temperatures [8-11]. Corrosion process causes destroy surface of material and creating corrosion products on surface of material. This factor can cause decrease magnetic properties of alloy [12-17].

In this paper the results of experience of influence a structure and a corrosion medium on the magnetic properties of  $\text{Fe}_{63.5}\text{Co}_{10}\text{Cu}_1\text{Nb}_3\text{Si}_{13.5}\text{B}_9$  alloy have been presented and compared with the achievements without influence of the corrosion.

## 2. Experimental

Researches include  $\text{Fe}_{63.5}\text{Co}_{10}\text{Cu}_1\text{Nb}_3\text{Si}_{13.5}\text{B}_9$  alloy in state „as quenched” obtained in the form of strips (thickness of about 25  $\mu\text{m}$ ) by the method of rapid cooling from liquid phase.

The strips of researched material in state „as quenched” were isothermally heat treated (denoted as an annealing temperature  $T_a$ ) in range 473-1023 K with the step 50 K, for 1 h. In a temperature 773-873 K when was expected the best magnetic permeability  $\mu$  with the step 25 K. The samples were annealing in an electrical chamber furnace Thermolyne Model No.F 6020C-60 with an argon atmosphere.

A measurement volume of magnetisation in function of the annealing temperature was made to define temperature of the primary crystallisation and the Curie temperature. The measurements used 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 constant speed of heating 5 K/min.

Results of measurements the force which researched sample was „sucked” or „upholster” from magnetic in define time during annealing allowed defining a ratio of magnetisation sample (volume magnetic phase in researched material).

A structural researches of strip were made using the X-ray diffractometer XRD 7 of firm Seifert-FPM allowed defining a structure of the material and participation crystal phase  $\alpha$ -(Fe,Co,Si) in alloy after the crystallisation dependently from the annealing temperature.

A Scherrer formula was used to defined size of granule in a crystal phase in material.

The corrosion researches were made by exposition the alloy by 14 days in 1 M corrosion medium NaCl. Then magnetic properties (initial magnetic permeability  $\mu_p$ , relative magnetic permeability  $\mu_w$ , coercive field  $H_c$ , remanence  $B_r$  and saturation magnetisation  $B_s$ ) were measured what allowed comparing measurements with results obtained from the alloy before influence the corrosion environment.

Initial magnetic permeability  $\mu_p$  designed 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 oscillator acoustic.

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

The other magnetic properties (coercive field  $H_c$ , remanence  $B_r$  and saturation magnetisation  $B_s$ ) measured using apparatus the VSM – Vibrating Sample Magnetometer.

In regard to big influence the surface of material on magnetic properties of alloy. The research also contains a roughness of the strip from both sides.

A mat surface had a contact with a cooling roll and with opposite side is glossy surface. The research of roughness worked out before and after influence corrosion medium in temperature  $T_{op}$ -798 K.

The fractography research of surface the samples from both sides were worked out for amorphous structure after relaxation and for alloy after the primary crystallisation.

## 3. Results and discussion

Results of measurements of magnetisation ratio in function of annealing temperature  $\text{Fe}_{63.5}\text{Co}_{10}\text{Cu}_1\text{Nb}_3\text{Si}_{13.5}\text{B}_9$  alloy allowed determining the temperature of primary crystallisation  $T_x$  and the Curie temperature  $T_c^{am}$ , fig. 1, 2.

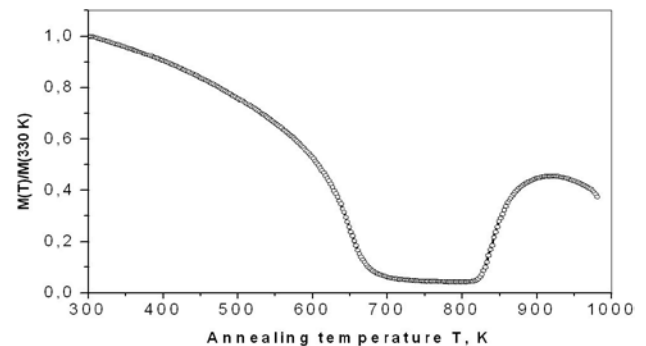


Fig. 1. Normalised curves of magnetisation determined in a function of annealing temperature for  $\text{Fe}_{63.5}\text{Co}_{10}\text{Cu}_1\text{Nb}_3\text{Si}_{13.5}\text{B}_9$  alloy

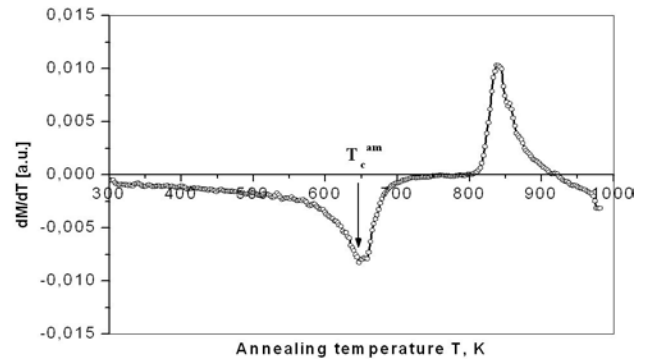


Fig. 2. Dependence  $dM/dT$  for normalised curves of magnetisation determined in function of annealing temperature for  $\text{Fe}_{63.5}\text{Co}_{10}\text{Cu}_1\text{Nb}_3\text{Si}_{13.5}\text{B}_9$  alloy

On the beginning of annealing the samples in range of temperatures 300-700 K the magnetisation of amorphous phase is decreasing. It is connected with transformation phase ferromagnetic – paramagnetic, getting near null, fig. 1. This point respond to minimal value of  $\frac{dM}{dT}$  (denoted as  $T_c^{am}$ ) and also is

noted as the Curie temperature, fig. 2. The Curie temperature for researched  $\text{Fe}_{63.5}\text{Co}_{10}\text{Cu}_1\text{Nb}_3\text{Si}_{13.5}\text{B}_9$  alloy is  $T_c^{am}$  - 646 K.

A low magnetisation of material in paramagnetic phase is until moment as starts the crystallisation amorphous phase. Formation of the new ferromagnetic phase in amorphous matrix gives increase of value of magnetisation in higher temperature of annealing. The temperature  $T_x$  817 K is treated as a temperature of primary crystallisation, fig. 1.

The research using the X-ray diffraction allowed defining quality changes in material structure caused by annealing alloy in the state „as quenched”. Also a degree of the crystallisation after the primary crystallisation was appreciated.

The X-ray diffraction of alloy defined that the strip of material obtained by the method of rapid cooling from liquid phase has the amorphous structure – „as quenched”. On a diffraction pattern is observed one characteristic widened a diffraction line in a place when is observed a diffraction line from plane (110)  $\alpha$ -Fe, fig. 3a. For the material after annealing in range of temperature 473-798 K the diffraction pattern doesn't demonstrated 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)  $\alpha$ -Fe, fig. 3b,c. The heat treatment in a temperature 823 K causes to observe the diffraction pattern peak from the crystal phase  $\alpha$ -(Fe,Co,Si) characteristic for the primary crystallisation, fig. 3d. This structure is characteristic for the crystal phase with amorphous matrix. The size of granule  $d$  – crystal phase defined using Scherrera formula for investigated  $\text{Fe}_{63.5}\text{Co}_{10}\text{Cu}_1\text{Nb}_3\text{Si}_{13.5}\text{B}_9$  alloy is  $d$  – 12.3 nm. The annealing researched alloy in temperature 923 K causes to perform the second stage of crystallisation. It is connected with changes in structure of alloy and compounds of borides of iron  $\text{Fe}_2\text{B}$  are observed, fig. 3e. The size of granule has changed and in this temperature is  $d$  – 14.4 nm, what is connected with decrease of amorphous matrix in the investigated strip. A full crystallisation of alloy is observed in a temperature 1023 K. This causes a bigger volume fraction of the compounds of borides  $\text{Fe}_2\text{B}$  and  $\text{Fe}_3\text{B}$ , fig. 3f. This processes caused a grain growth of alloy to a value  $d$  – 35 nm.

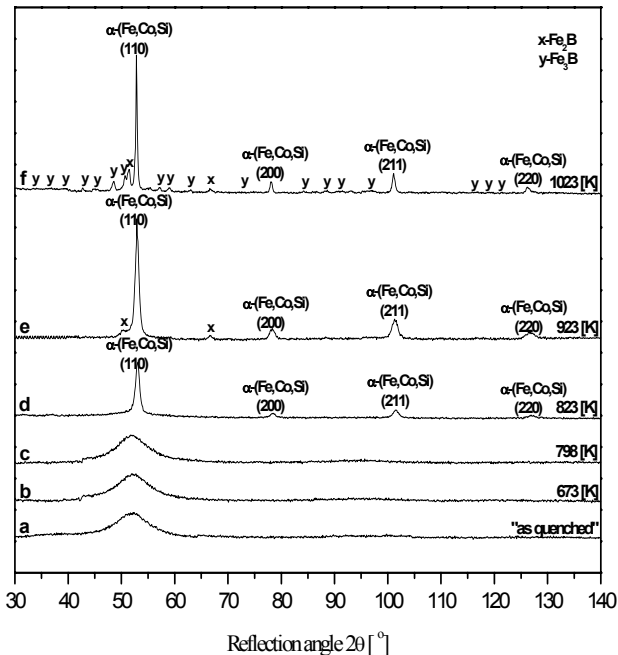


Fig. 3. X-ray diffractions investigated  $\text{Fe}_{63.5}\text{Co}_{10}\text{Cu}_1\text{Nb}_3\text{Si}_{13.5}\text{B}_9$  alloy in state „as quenched” and after annealing

A estimation of participation the crystal phase  $\alpha$ -(Fe,Co,Si) in all volume of the alloy for a different temperature of annealing allow observing that with increasing of the temperature the volume of crystal phase  $\alpha$ -(Fe,Co,Si) is also increasing. In the temperature 823 K a volume of crystal phase amounts 45 % for  $\text{Fe}_{63.5}\text{Co}_{10}\text{Cu}_1\text{Nb}_3\text{Si}_{13.5}\text{B}_9$  alloy. Increasing the heat

treatment to the temperature 923 K causes increasing a volume of crystal phase  $\alpha$ -(Fe,Co,Si) to volume 68 % what is connected with noted the compounds of borides  $\text{Fe}_2\text{B}$ . The alloy in temperature 1023 K has about 85 % the crystal phase  $\alpha$ -(Fe,Co,Si) and the compounds of borides  $\text{Fe}_2\text{B}$  and  $\text{Fe}_3\text{B}$ .

Initial magnetic permeability  $\mu_p$  of  $\text{Fe}_{63.5}\text{Co}_{10}\text{Cu}_1\text{Nb}_3\text{Si}_{13.5}\text{B}_9$  alloy changes with change of structure of material in function of annealing temperature. Increasing of value of the permeability is observed up to maximum value in a temperature 798 K, fig. 4. The exposure the material in corrosion medium 1 M NaCl by 14 days had influence on decreasing of the initial magnetic permeability  $\mu_p$ . In the temperature 798 K maximal value of initial permeability is 5258, what suggests scientifically decrease of the permeability after influence of the corrosion, fig. 4.

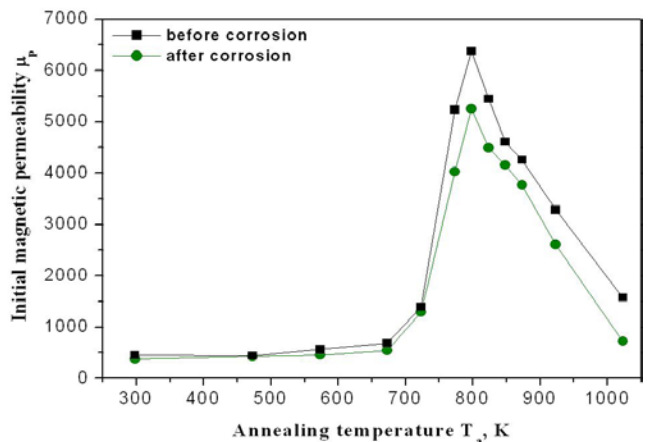


Fig. 4. Initial magnetic permeability  $\mu_p$  for  $\text{Fe}_{63.5}\text{Co}_{10}\text{Cu}_1\text{Nb}_3\text{Si}_{13.5}\text{B}_9$  alloy as a function of annealing temperature in field 0.5 A/m, before and after exposure in 1 M NaCl corrosion medium

The  $\text{Fe}_{63.5}\text{Co}_{10}\text{Cu}_1\text{Nb}_3\text{Si}_{13.5}\text{B}_9$  alloy before and after exposure in 1 M NaCl corrosion medium has the best initial magnetic permeability  $\mu_p$  in the temperature 798 K, denoted as a optima temperature  $T_{op}$ . The researches indicate on significant decreasing the permeability after exposure in 1 M NaCl corrosion medium. The decrease of the magnetic properties can be caused by formation the compounds of corrosion on the surface of the strips.

The researches of the relative magnetic permeability  $\mu_w$  indicate that heat treatment and the magnetic field  $H$  have a big influence on the permeability. The material after annealing in temperature in  $T_{opt}$  798 K characterises a fivefold increasing the relative permeability in relation to the material in the state „as quenched”, fig. 5.

The results of research the relative magnetic permeability  $\mu_w$  also indicate on decrease the magnetic properties of the alloy after exposure in 1 M NaCl corrosion medium, fig. 5. A decrease the value of relative permeability and a shift a maximum to higher magnetic field for state „as quenched” and after annealing is observed.

The relative magnetic permeability for the  $\text{Fe}_{63.5}\text{Co}_{10}\text{Cu}_1\text{Nb}_3\text{Si}_{13.5}\text{B}_9$  alloy indicates on significant influence the corrosion medium on the magnetic properties. It can be caused by deposition products of the corrosion on the surface of material.

The material in state „as quenched” after annealing indicates a change of the magnetic properties (coercive field  $H_c$ , remanence  $B_r$  and saturation magnetisation  $B_s$ ). The isothermal annealing in temperature 673 K has influence on improving the coercive field  $H_c$ . The increase of the temperature of annealing to value  $T_{opt}$ -798 K responds to the best initial magnetic permeability and the structural relaxation causes to improve the coercive field  $H_c$ , remanence  $B_r$  and saturation magnetisation  $B_s$ . The crystallisation process of alloy in higher temperatures has influence on decreases the coercive field  $H_c$ . Influence of 1 M NaCl corrosion medium is also observed for the magnetic properties as: coercive field  $H_c$ , remanence  $B_r$  and saturation magnetisation  $B_s$ . It is also observed influence of corrosion on magnetic properties  $Fe_{63.5}Co_{10}Cu_1Nb_3Si_{13.5}B_9$  alloy. The annealing had influence on increase of susceptibility of the material on the corrosion medium. The most significant decrease of the magnetic properties is observed for the alloy after crystallisation.

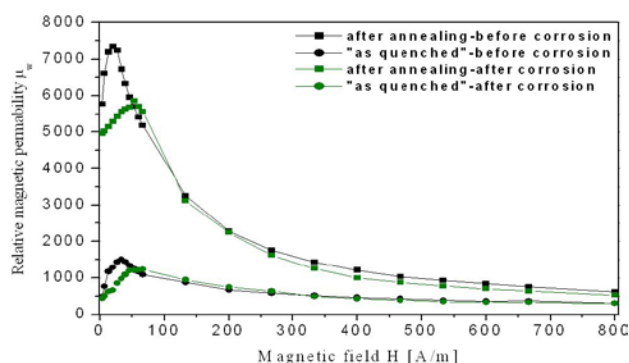


Fig. 5. Relative magnetic permeability  $\mu_w$  for  $Fe_{63.5}Co_{10}Cu_1Nb_3Si_{13.5}B_9$  alloy as a function of magnetic field for samples in state „as quenched” and after annealing, before and after exposure in 1 M NaCl corrosion medium

## 4. Conclusions

- The results of the research  $Fe_{63.5}Co_{10}Cu_1Nb_3Si_{13.5}B_9$  alloy indicating on big dependence between the magnetic properties, the structure of material and influence of 1 M NaCl corrosion medium.
- The optimal magnetic properties of the strips were obtained for the amorphous structure after structural relaxation, before and after exposition in the corrosion medium.
- The coercive field  $H_c$  in the optimal temperature  $T_{opt}$ -798 K decreased after influence 1 M NaCl about 2 A/m

## References

- [1] A.C Sousa, C.S. Kiminami, Crystallization and corrosion of amorphous FeCuNbSiB, *Journal of Non-Crystalline Solids* 219 (1997) 155-159.
- [2] Ajay Gupta, Kailash Ruwali, Neelima Paul, P. Duhaj, Crystallization behavior of amorphous  $(Fe_{100-x}Co_x)_{85}B_{15}$ , *Materials Science and Engineering A* 304-306 (2001) 371-374.
- [3] T. Gloriant, S. Surinach, M.D. Baro, Stability and crystallization of Fe-Co-Nb-B amorphous alloys, *Journal of Non-Crystalline Solids* 333 (2004) 320-326.
- [4] J. Zbrozarczyk, J. Olszewski, W. Czurzyńska, B. Wysocki, R. Kolano, A. Młyńczyk, M. Łukiewski, A. Kolano, J. Lelątko, Microstructure and some magnetic characteristics of amorphous and nanocrystalline  $Fe_{83-x}Co_xNb_3B_{13}Cu_1$  ( $x=0$  or 41.5) alloys, *Journal of Magnetism and Magnetic Materials* 254-255 (2003) 513-515.
- [5] C.F. Conde, A. Conde, P. Svec, P. Ochín, Influence of the addition of Mn and Cu on the nanocrystallization process of HITPERM Fe-Co-Nb-B alloys, *Materials Science and Engineering A* 375-377 (2004) 718-721.
- [6] M.E. McHenry, M.A. Willard, D.E. Laughlin, Amorphous and nanocrystalline materials for applications as soft magnets, *Prog. Mater. Sci.* 44 (1999) 291-433.
- [7] J.S. Blazquez, V. Franco, A. Conde, M.R.J. Gibbs, H.A. Davies, Z.C. Wang, The evolution of magnetostriction and coercivity with temperature in the early stages of nanocrystallisation in FeCoNbB(Cu) alloys *Journal of Magnetism and Magnetic Materials* 250 (2002) 260-266.
- [8] A. Kolano-Burian, R. Kolano, M. Polak, Microstructure and soft magnetic properties of Finemet alloys modified by Co, *CAM3S* (2005) 488-491.
- [9] A. Altube, H. Takenouti, L. Beaunier, M. Keddad, S. Joiret, S. Borensztajn, F. Pillier, A.R. Pierna, A microscopic and impedance spectroscopy study of Finemet-Co amorphous alloys, *Corrosion Science* 45 (2003) 685-692.
- [10] V. Cremaschi, A. Saad, M.J. Ramos, H. Sirkin, Magnetic and structural characterization of Finemet type alloys with addition of Ge and Co, *Journal of Alloys and Compounds* 369 (2004) 101-104.
- [11] S. Lesz, R. Nowosielski, B. Kostrubiec, Z. Stokłosa, Crystallization kinetics and magnetic properties of a Co-based amorphous alloy, *CAM3S* (2005) 574-577.
- [12] D. Szewieczek, A. Baron, G. Nawrat, Protect before corrosion (2005) Nr 6, 232-237, (in Polish).
- [13] D. Szewieczek, A. Baron, Electrochemical corrosion properties of amorphous  $Fe_{78}Si_{13}B_9$  alloy, *Journal of Materials Processing Technology* 157-158 (2004) 442-445.
- [14] A. Baron, D. Szewieczek, G. Nawrat, Studies of amorphous and nanocrystalline  $Fe_{73.5}Si_{13.5}B_9Nb_3Cu_1$  alloy corrosion: Electrochemical impedance spectroscopy under polarization condition, *CAM3S* (2005) 66-71.
- [15] D. Szewieczek, A. Baron, Electrochemical corrosion and its influence on magnetic properties of  $Fe_{75.5}Si_{13.5}B_9Nb_3Cu_1$  alloy, *AMME* (2005) 631-636.
- [16] N. A. Mariano, C. A. C. Souza, J. E. May, S. E. Kuri, Influence of Nb content on the corrosion resistance and saturation magnetic density of FeCuNbSiB alloys, *Materials Science and Engineering A* (2003) 1-5.
- [17] C. A. C. Souza, J. E. May, L. Bolfarini, S. E. Kuri, M. F. de Oliveir Cremaschi, J. Osewski, C. S. Kiminami, Influence of composition and partial crystallization on corrosion resistance of amorphous Fe-M-B-Cu ( $M=Zr, Nb, Mo$ ) alloys, *Journal of Non-Crystalline Solids* 284 (2001) 99-104.