Journal of Achievements in Materials and Manufacturing Engineering
VOLUME 14
ISSUE 1-2
January-February
2006

Manufacturing technology of the composite materials: nanocrystalline material – polymer type

B. Ziębowicz, D. Szewieczek, L.A. Dobrzański*
Division of Materials Processing Technology and Computer Techniques in Materials Science, Institute of Engineering Materials and Biomaterials,
Silesian University of Technology, ul. Konarskiego 18a, 44-100 Gliwice, Poland
* Corresponding author: E-mail address: leszek.dobrzanski@polsl.pl

Received 15.11.2005; accepted in revised form 31.12.2005

Materials

ABSTRACT

Purpose: This paper presents the material and technological solution which makes it possible to obtain the nanocrystalline, ferromagnetic powder material of $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ alloy after its thermal nanocrystallization with the succeeding high-energy milling. Another aspect was to develop the technology to obtain the nanocrystalline composite materials made by binding the obtained powder material with the high density low-pressures polyethylene (PEHD) with the controlled ferromagnetic and mechanical properties.
Design/methodology/approach: Magnetic properties of composite materials were examined on the Lake Shore Cryotronics Inc VSM vibratory magnetometer. Compression strength of composite materials was determined on Instron 1195 universal testing machine.
Findings: The research carried out has made it possible to develop the general technological process that makes it possible to fabricate composite materials consisting of the nanocrystalline powders of the soft magnetic material and thermoplastic polymer with the required magnetic and mechanical properties. Examination of mechanical properties show that these materials have satisfactory compression strength. Composite materials show lower magnetic properties in comparison with magnetic powder but their geometrical form allows to extend their applications.
Practical implications: The usability of composite materials nanocrystalline material – polymer type as inductive component in electronic industry depends upon further investigations.
Originality/value: Soft magnetic composite materials based on $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ alloy powders are essential elements in today's electronic world. As far as we know, composites materials based on nanocrystalline material as soft magnetic materials have not been examined yet.
Keywords: Composites; Nanomaterials; FINEMET alloy; High density low-pressures polyethylene; One-sided uniaxial pressing

1. Introduction

The observed in the recent years progress in the magnetic materials engineering referred mainly to the good mastery of manufacturing technology of materials with amorphous and nanocrystalline structure [1-8]. In case of these materials, their structure is a decisive factor determining new quality properties in comparison to the conventional properties of crystal materials. Unfortunately, amorphous and nanocrystalline materials apart from much better magnetic properties, have limitations concerning their application because of their geometrical form –

powder or thin, brittle tape, 0.02-0.05 thick – that results from most often used methods of their manufacturing (fast cooling of liquid crystal connected with a controlled crystallization and mechanical synthesis). In order to obtain a solid material with appropriate resistant properties and thereby widen the application possibilities, one needs to use the bonding of the received powders and crumbled tapes with the polymer, fusible metal or hot moulding. The application of nonmagnetic binding factor brings about the fact that thus obtained composite material has worse magnetic properties in comparison to its magnetic component [9-16].

There are many publications whose autors deal with the problems of manufacturing the nanocrystalline magnetic materials and their properties [1-8]. Unfortunately, there are no literature sources which would refer in a satisfactory way to consolidation of the magnetic soft nanomaterials with various materials, including polymers, and to the mechanical and physical properties of the composite materials obtained in this way.

There are complex relationships among the manufacturing technology of these materials, their microstructure, as well as their mechanical and physical properties, which pose one of the still unsolved satisfactorily matters of interest of the materials engineering.

This paper presents the material and technological solution leading to obtaining the composite materials consisting of the nanocrystalline powders of the $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ alloy and polyethylene. The technological concept is based on the use of the process of thermal nanocrystallization of amorphous tapes and, next, their high-energy milling in order to produce a powder with an optimal particle quantity, and binding them with polyethylene into composite material. Having realized the proposed technological concept, the composite materials have been manufactured as a result of one-sided and uniaxial moulding of the prepared components.

2. Components

To manufacture a composite material a tape from a metallic glass in 'as quenched' state has been used with a chemical composition and geometrical features as given in Table 1 as well as low-pressure polyethylene with a big density (PEHD) – Table 2.

- The strips from the investigated alloy were cast with the planar-low-casting (PFC) onto the surface of the rotating cooling drum in the argon protective atmosphere. The chemical composition of the tapes has been determined using JEOL-MICROPROBE 733 electron microanalyzer with energodispersion attachment of LINK company. The heat treatment (thermal nanocrystallization) of strips in the as quenched state was carried out in the Thermolyne 6020C chamber electric furnace with the electronic temperature measurement and control, in the argon protective atmosphere, in the sealed container, through which argon was flowing at constant rate. Temperature regulation range was ±2K. The strips for the heat treatment were coiled into the toroidal cores with the outer diameter D=30 mm, inner diameter d=25mm, and height of h=9mm.
  The strips were heat treated by holding at the temperature of 823 K for 1 h in the argon atmosphere (thermal nanocrystallization). Phases: crystalline αFe(Si) and the amorphous intergranular one - developed as a result of the primary crystallization, are the products of heat treatment of the $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ metallic glass strips. The nanocrystallization temperature selected in this way ensures the most advantageous magnetic properties for this alloy.
  Powders of the $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ alloy after its thermal nanocrystallization were made by the high-energy grinding of the initially crumbled strips in the shaker type 8000 SPEX CertiPrep Mixer/Mill for 0.25, 1.0, 3.0, 5.0, 10.0 and 20.0 hours in the air (Fig. 1).

a)

b)

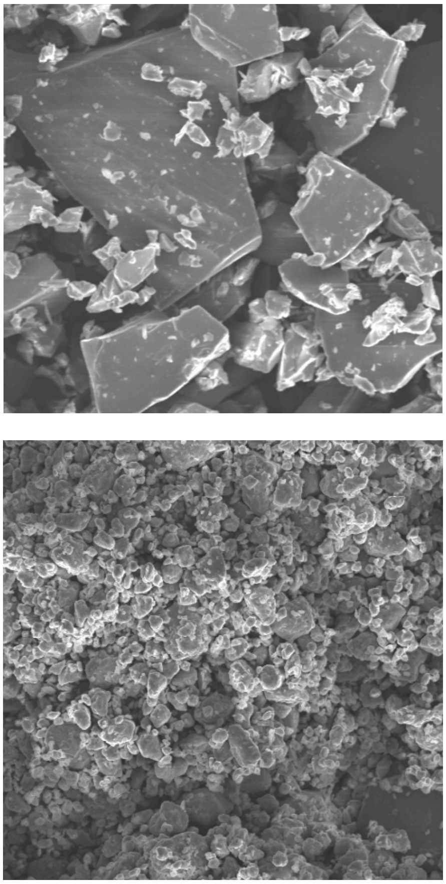


Fig. 1. The shape of $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ powder particle – milling time: a) 0.25 h, b) 20.0 h (OPTON DSM scanning electron microscope - magnification 500x)

  In this shaker, the container with the diameter of ϕ = 38 mm and height h = 63mm with balls (four steel balls with the diameter of ϕ =6.3 mm and two steel balls with the diameter of ϕ =12.7mm) and the ground material generates vibrations of the grinding medium and material inside it, during which their collisions occur. Only 15g of material was ground at one time. The strips were reeled off the toroidal cores before grinding and cut initially into segments about 10 mm long each.
- The high density low-pressure polyethylene (PEHD) powder was used as binder – with mass fractions of 1.0%, 2.5%, 5.0%, 7.5% and 10.0%. The powder was manufactured by ASHLAND POLAND company (Table 2, Fig. 2).

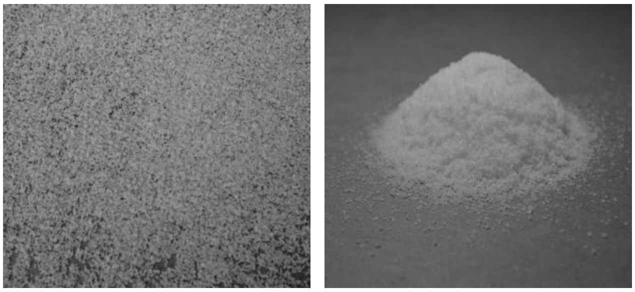


Fig. 2. View of the PEHD powder particle

- The choice of this material has been made because it is the cheapest and most often used thermal polymer, easy in processing, and its processing temperature of 443K causes at the same time the relaxation of internal stresses arisen as a result of milling of $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ alloy tapes. This simplifies the technology of manufacturing of composite materials through combining in one operation the processes of relaxation of internal stresses in powders and the process of moulding. It has been assumed that the polyethylene mass participation in the composite material should be minimized considering that it is a nonmagnetic material and should assure its coherence.

3. Obtaining of composite materials

The composite materials have been obtained as a result of one-sided anaxial moulding of earlier prepared components. For this purpose a module for composite forming has been designed consisting of the die and the heater enclosed which has been coupled with Zwick Z100 strength machine set up for compression (Fig. 3). The moulding took place in atmospheric pressure.

The choice of the method of obtaining composites determines the following criteria:

- this method enables to make any elements with sophisticated shapes,
- the process of moulding has been executed at the 443K temperature what is important considering metastable structure of $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ powders,
- during the moulding process the $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ alloy powder particles get closer to each other what increases their contact area and causes their mechanical meshing.

The times of high-energetic $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ alloy tapes milling have been determined in the following way:

- the times which least of all generate internal stresses that cause the lowering of magnetic properties of the obtained powders have been accepted,
- the times after which obtained the differentiation of shapes and quantity of particles of powders have been accepted.

For manufacturing composite materials $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ alloy powders have been chosen arisen as a result of type milling for 0.25, 1.0, 3.0 and 5.0 hours.

Among the examined polyethylene mass portions in composite materials the biggest one is 7.5% as bigger participation of this component in the composite material causes its flowing out of the die while moulding. The smallest polyethylene mass portion is 2.5% as the smaller participation of the component does not ensure its cohesion. In other examinations 2.5, 5.0 and 7.5% polyethylene mass portions in composite materials have been employed.

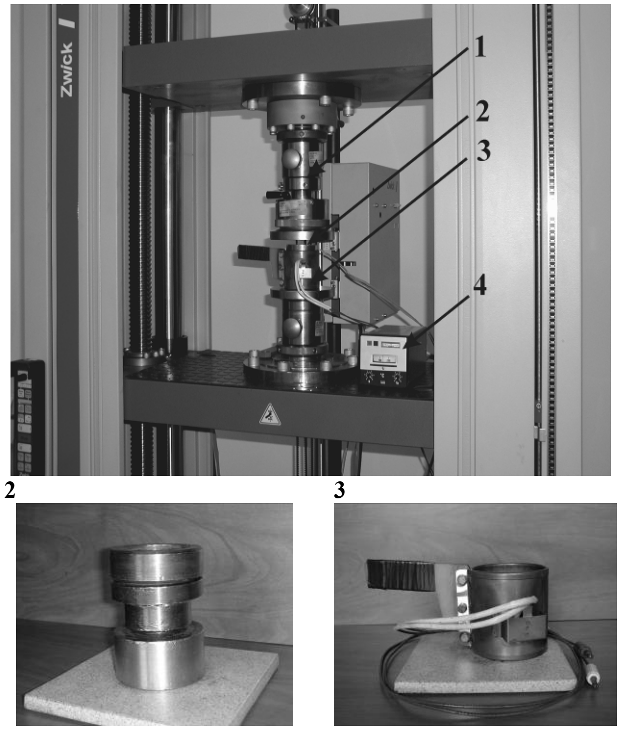


Fig. 3. View of the composite material forming stand: (1) testing machine set up for compression; (2) die block, (3) heater, and (4) thermometer

The components for manufacturing composite materials have been mixed in the 8000 SPEX CertiPrepMixer/Mill shaker. Time of mixing was 0.10, 0.25, 0.50 h.

To obtain densities of composite materials the most similar to the theoretical density, the most possible for implementation 350MPa moulding pressure have been chosen. The obtaining of the possibly biggest density diminishes air cracks in composite materials. The growth of air-gaps causes the linearization of interrelations B=f (H) and what follows the deterioration of magnetic properties of manufactured materials.

Among the applied times of the components' mixing, the 0.25 h time is optimal as it is the shortest of applied times which allows for adhesion of $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ alloy particles to polyethylene particles.

Among the applied moulding times, 0.25 h is optimal as it allows for the fastest, entire binding of the used components into the composite material.

The material-technological research done allows to prepare a frame technological process of receiving composites consisting of nanocrystalline powders of a magnetic soft material and thermal polymer with the most favourable combination of mechanical and magnetic properties (Fig. 5).

Table 1
Chemical constitution and dimensions of amorphous tapes used to manufacturing composite materials

Designation		$Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$
Brand name		FINEMET
Concentration [%]	Fe	73.5
	Si	13.5
	B	9.0
	Nb	3.0
	Cu	1.0
Dimensions [mm]	thickness	0.03
	width	9.0

Table 2
Characteristic of polyethylene used to manufacturing composite materials [10]

Characteristic	Description
Geometrical form	powder
Grain size [µm]	50÷650
Density ρ [g/cm^3]	0.94
Shore hardness	55
Stretching R_m [MPa]	20
Processing temperature [K]	433÷523

Table 3
Properties of composite materials with the most favourable combination of mechanical and magnetic properties

Properties		Composite material: PEHD 5% mass portion + $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ alloy powder obtained by milling the strips for 0.25 h.
mechanical	Compression strength R_c [MPa]	50.25
	Unit shortening A_c [%]	10.34
	Young's moduluj E_c [MPa]	485.75
magnetic	Coercive force intensity H_c [A/m]	421.88
	Remanencie B_r [T]	0.0034
	Saturation induction B_s [T]	1.248
	Permeability μ_{max}	80
	Maximum power losses P_{max} [W/kg]	1.804

Among the manufactured materials, the best mechanical properties show the composite materials with 5.0 % mass fraction of polyethylene (Fig.4, Table 3).

Magnetic properties of composite materials were examined on the Lake Shore Cryotronics Inc VSM vibratory magnetometer with the working voltage of 30 V, maximum field intensity 1800 kA/m and the time-constant 3 s. The examination results were collected and processed using the IDEAS^{TM} VSM Software package which featured the integral part of the VSM system.

Compression strength of composite materials was determined on Instron 1195 universal testing machine. Compression rate was 5 mm/min. The test was carried out at room temperature. Three tests were made for each composite version.

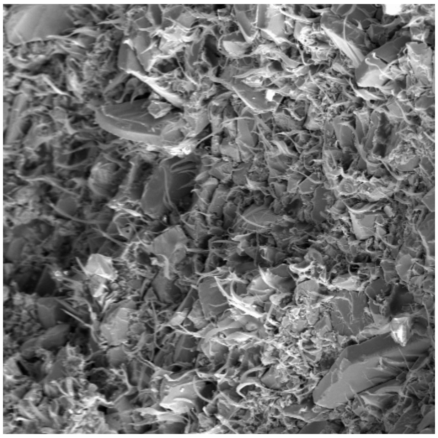
Such a quantity of polyethylene in the composite material allows to approach to each other the powder particles. It also allows to increase their contact surface and their reciprocal meshing what has made the operation of the adhesive force possible. For the composite materials with a smaller mass portion of polyethylene these phenomena are also true but the quantity of polyethylene is insufficient for binding all powder particles of $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ alloy. For the composite materials with a bigger polyethylene mass portion the reciprocal meshing of particles and their contact surface are smaller considering larger quantities of polyethylene which limits these phenomena.

The best mechanical properties possess the composite materials with nanocrystalline powder of $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$

Table 4
The characteristics of composite materials with the best combination of mechanical and magnetic properties

Characteristic	Description
Polyethylene mass fraction [%]	5
Milling time of used $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ alloy [h]	0.25
Average grain size of used $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ alloy [μm]	34.71
Theoretical density ρ_t [g/cm^3]	7.36
Average density ρ_a [g/cm^3]	4.72

a)



b)

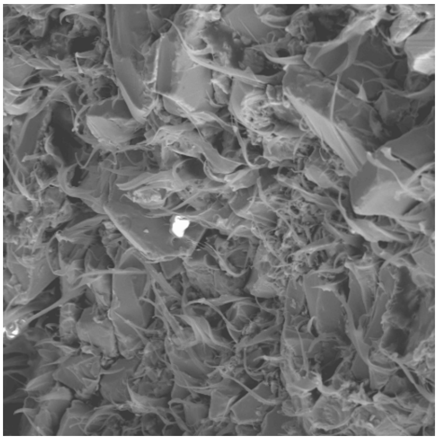


Fig. 4. Microscopic image of composite material fractures obtained after decohesion in the compression test – PEHD mass portion of 5.0% with powder $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ alloy – milling time 0.25h – a) magnification 250x, b) magnification 500x

alloy that emerged from tape milling at the time of 0.25 h (Table 3). These materials show the least values of coercive force field intensity and magnetic residue as well as the highest values of relative permeability and saturation induction. Shape and size of particles of the powders used has the biggest influence on the magnetic properties of composites. Particles obtained by milling strips from the $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ alloy for 0.25 h are the biggest and have the shape of lamellar with sharp edges and of elongated flakes, which causes that the demagnetization phenomenon is the smallest for these particles (Fig. 1, Table 4). The polyethylene mass portion in the composite and the internal structure of the $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ alloy powders decides their magnetic properties to a less extent.

The characteristics of composite materials with the best combination of mechanical and magnetic properties has been presented in Table 4.

The density measurements of the test pieces of the obtained composite materials were carried out by determining the test piece mass using the analytical balance with the accuracy of 0.001g and its volume basing on the apparent mass loss by immersing in water. Five measurements were made for each composite version.

The obtained composite materials show worse magnetic properties in comparison to the magnetic properties of tapes and $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ alloy powders [9-13]. However, their value of magnetic permeability of about 80, satisfactory mechanical properties and the possibility of obtaining any, sophisticated shapes, make them very useful. Their price and quality is also crucial as there are many other materials with similar magnetic properties. The presented in the paper material-technological solution shows that the obtained composite materials can be used as the elements of computer and telephone exchange feeders.

4. Summary

Basing on the investigations carried out it was found out that the most advantageous conjunction of the magnetic and mechanical properties is displayed by composites with the 5% polyethylene mass portion with the nanocrystalline $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ alloy powder obtained by milling the strips for 0.25 h.

The worked out, frame technological process of composite materials consisting of nanocrystalline powders of magnetically soft material and thermal polymer may present the basis for applying different technological modifications that make it possible to choose any operations in terms of the expected mechanical and magnetic properties of composite materials.

The manufacturing of composite materials greatly expand the applicable possibilities of nanocrystalline powders of magnetically soft materials.

References

[1] J.Hu, H. Qin, S. Zhou, Y. Wang, Z. Wang, Materials Science and Engineering, B83 (2001) 24.

[2] A. Hernando, A. Gonzales, A. Salcedo, F. J. Palomares, J. M. Gonzales, Journal of Magnetism and Magnetic Materials, 221 (2000) 172.

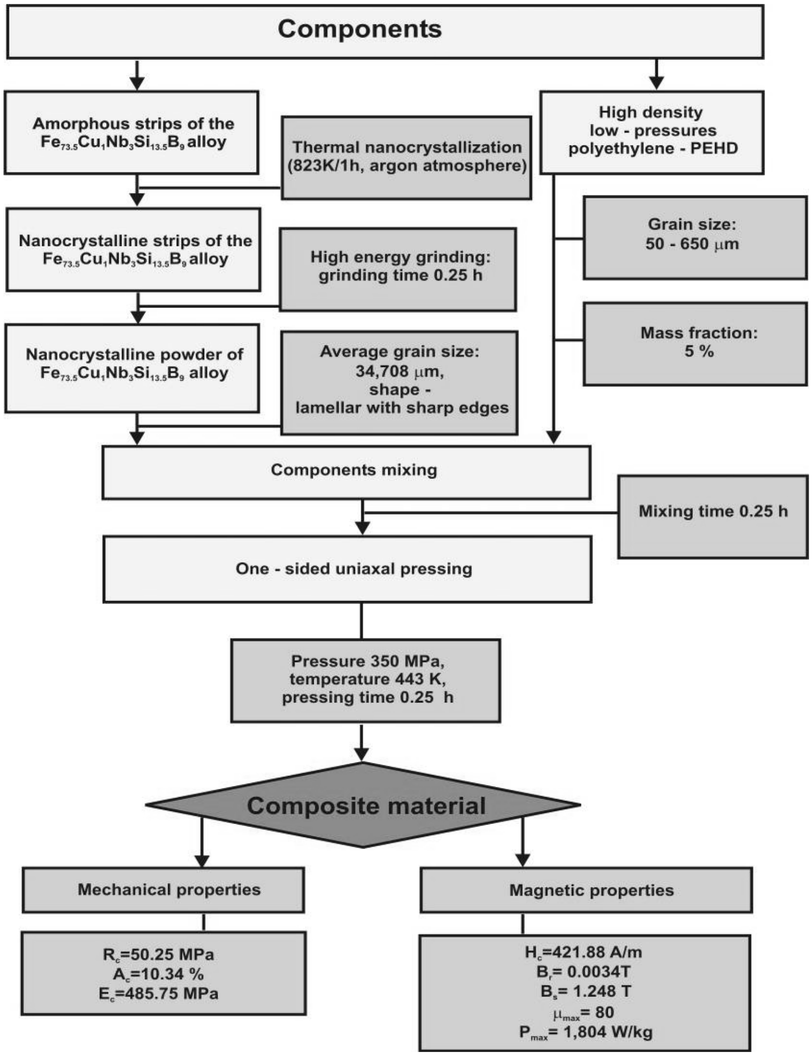


Fig. 5. Schematic diagram of the materials and technological operations for manufacturing the composite material with the most advantage combination of its mechanical and magnetic properties

[3] M. E. McHenry, M. A. Willard, D. E. Laughlin, Progress in Materials Science. 44 (1999) 91.
[4] Y. Yoshizawa, Journal of Metastable and Nanocrystalline Materials, 1 (1999) 51.
[5] E. Fechova, J. Kovač, P. Kollar, J. Füzer, M. Konč, Materials Science Forum, 360-362 (2001) 557.
[6] Z. Stokłosa, J. Rasek, P. Kwapuliński, G. Haneczok, G. Badura, J. Lelątko, Materiale Science and Engineering C23 (2003) 49.
[7] T. Kulik, Journal of Non-Crystalline Solids 287 (2001) 145.
[8] C. Suryanarayana: Progress in Materials Science, 46 (2001) 1.
[9] D. Szewieczek, L. A. Dobrzański, B. Ziębowicz, Journal of Materials Processing Technology, 157-158 (2004) 765.
[10] B. Ziębowicz, Structure and properties of the composite materials consisting of the nanocrystalline $Fe_{73.5}Cu_1Nb_3Si_{13.5}B_9$ alloy powders and polyethylene, PhD thesis – unpublished, Main Library of the Silesian University of Technology, Gliwice 2004 (in Polish).
[11] B. Ziębowicz, D. Szewieczek, L. A. Dobrzański, Journal of Materials Processing Technology, 157-158 (2004) 776.
[12] B. Ziębowicz, D. Szewieczek, L. A. Dobrzański, Proceedings 12th International Scientific Conference AMME'2003, Gliwice-Zakopane, 2003, 1077.
[13] B. Ziębowicz, D. Szewieczek, L.A. Dobrzański, J. J. Wysłocki, A. Przybył, Journal of Materials Processing Technology, 149-155, (2005) 149.
[14] B. Ziębowicz, D. Szewieczek, L. A. Dobrzański, Proceedings of the third scientific conference M3E'2005, Gliwice-Wisła, 2005, 261.
[15] L.A. Dobrzański, M. Drak, Journal of Materials Processing Technology, 157-158 (2004) 650.
[16] H. Chiriac, A. E. Moga, M. Urle., F. Necula, , Journal of Magnetism and Magnetic Materials, 203 (1999) 159.
[17] D. Nuetzel, G. Rieger, J. Wecker, J. Petzold, M. Mueller, Journal of Magnetism and Magnetic Materials, 196-197 (1999) 323.