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Influence of temperature on structure and magnetic properties of powders alloys

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Materials

ABSTRACT

Purpose: The paper presents the research results of nanocrystalline powders obtained by high energetic milling of amorphous ribbons based on cobalt $Co_{77}Si_{11,5}B_{11,5}$ and $Co_{68}Fe_4Mo_1Si_{13,5}B_{13,5}$.

Design/methodology/approach: A 8000 SPEX CertiPrep Mixer/Mill high energy ball mill was applied to mill the ribbons both in „as quenched" state and heat treated. Observations of the structure of powders were made on the Opton DSM-940 scanning electron microscope. The change of powder material structure was measured with electron transmitting microscope JEOL JEM 200CX and X-ray analysis. The X-ray tests were realized with the use of the XRD 7 SEIFERT-FPM diffractometer.

Findings: The analysis of the magnetic properties test results of the of the $Co_{77}Si_{11.5}B_{11.5}$ and $Co_{68}Fe_4Mo_1Si_{13,5}B_{13,5}$.powders obtained in the high-energy ball of milling process proved that the process causes significant decrease in the magnetic properties. The structure and magnetic properties of this material may be improved by means of a proper choice of parameters of this process as well as the final thermal treatment.

Research limitations/implications: For the powders, further magnetical, structure and composition examinations are planed.

Practical implications: The amorphous and nanocrystalline powders of $Co_{77}Si_{11,5}B_{11,5}$ and $Co_{68}Fe_4Mo_1Si_{13,5}B_{13,5}$. alloys obtained by high-energy ball milling of metallic glasses feature an alternative to solid alloys and make it possible to obtain the ferromagnetic nanocomposites, whose shape and dimensions can be freely formed.

Originality/value: The paper presents influence of annealing temperature and parameters of the high-energy ball milling process on structure and magnetic properties of soft magnetic powder materials obtained in this technique. Results and a discussion of the influence of high energy mechanical milling process on particle size and their distribution and annealing temperature of powders as well as structure and magnetic properties of investigated samples is presented. According to achieved results it has been attempted to describe the possibilities of improvement the soft magnetic properties of obtained powder materials.

Keywords: Nanomaterials; Powders; Heat treatment; Magnetic properties

1. Introduction

The amorphous and nanocrystalline materials arouse interest of scientists in research centres all over the world in the last tree decades. This is connected with the soft magnetic properties characteristic for these materials and with the possibility of using them in the electronic and electrical industry [1-9].

The metallic amorphous and nanocrystalline materials obtained by crystallization of the metallic glasses are available in the form of thin ribbons only, and their magnetic properties can be controlled by heat- or thermo-magnetic treatment. These limitations are complicated in addition by the feasibility of manufacturing only toroidal cores from the ribbons.

Obtaining the nanocrystalline metal powders by milling of metallic glasses features an alternative to solid materials like amorphous and nanocrystalline ribbons and makes it possible to obtain the ferromagnetic nanocomposites, whose shape and dimensions can be freely formed using various consolidation methods [10-15].

2. Material and methods

The investigations were carried out on a $Co_{77}Si_{11,5}B_{11,5}$ and $Co_{68}Fe_4Mo_1Si_{13,5}B_{13,5}$ metallic glass in form of 0.036 mm thick and 10.2 mm wide ribbons. A 8000 SPEX CertiPrep Mixer/Mill high energy ball mill was applied to mill the ribbons both in „as quenched" state and heat treated. The vibration times were 5, 10, 15, 20 and 25 hours. A THERMOLYNE F6020C resistance furnace was used for isothermal soaking of the amorphous ribbons and the powder. Soaking was carried out at temperature range 300÷600^{o}C with 100^{o}C step in the argon atmosphere.

The X-ray tests were realized with the use of the XRD 7 SEIFERT-FPM diffractometer equipped with the lamp of the cobalt anode of 35 kV voltage and 30 mA filament current was used. Diffraction tests were carried out in the 2θ angle range from 40 to 120° (measurement step 0,1^o). Pulse counting time was 5 s.

Microscope examinations were made under the OPTON DSM 940 electron scanning microscope and the JEOL JEM 200CX electron transmision one. Tests of magnetic properties were carried out by the use of Lake Shore's Vibrating Sample Magnetometer VSM model 7307.

A MICROCAL ORIGIN 6.0 programme was used for the graphical analysis of the obtained X-ray photographs and $H_c=f(T_A)$ relation.

3. Results and discussion

The observation of materials in the scanning microscope proved that the high energy ball milling of the amorphous $Co_{77}Si_{11,5}B_{11,5}$ ribbon lasting 5 hours, results in the production of flakes of ribbons ("scales"). The shape of powder grains changes together with the time of milling. After 5-hour milling the shape of grains resembles flakes and after 15-hour milling the grains are smaller and more spherical (Fig. 1).

It was revealed, basing on magnetic examinations of powder material, that the coercion field value increases throughout the entire high energy milling time span and reaches its max. of H_C= 2610 A/m for the powder obtained after 25 hours of milling (Fig. 2).

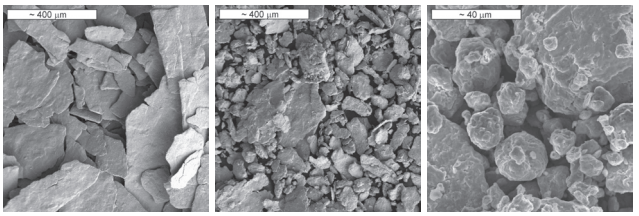

Fig. 1. Powder $Co_{77}Si_{11,5}B_{11,5}$ grains image after a) 5 h, b) 15 h, c) 30 h of the high energy miilling, scanning microscope

The saturation induction B_S value grows also along with the milling time. The B_S value for the powder obtained after 5 hours of milling is 0.63 T.

The powder obtained after 20 hours of the high energy milling characterized by the saturation induction of B_S=0.74 T. The B_S value increases as the milling process continues - like the coercion field value, reaching 0.77 T after 25 hours of milling. Increase of the coercion field H_C and of the saturation induction B_S takes place at the further stage of the process, as the milling time gets longer.

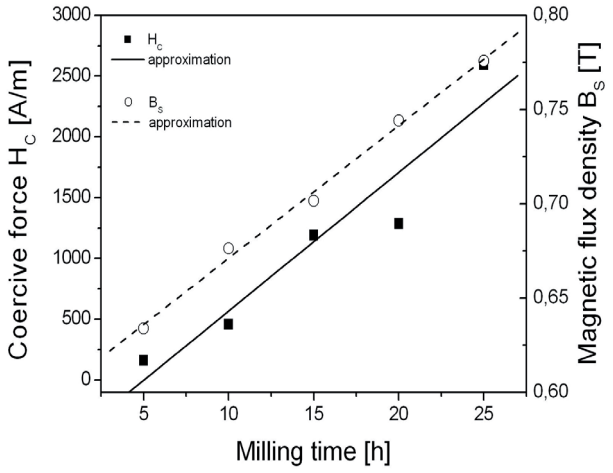

Fig. 2. Coercion field and saturation induction versus milling time of powders obtained from the $Co_{68}Fe_4Mo_1Si_{13,5}B_{13,5}$ amorphous ribbons

The magnetic research of the $Co_{77}Si_{11,5}B_{11,5}$ powders obtained in the process of milling of the ribbons in the "as quenched" state proved that the process of the high energy ball milling causes significant increase in the coercive force. The powder obtained after 5-hour milling of the amorphous ribbon is characterised by the highest value of the coercive force field (H_C=517,1). The longer the time of milling is, the higher the value of the parameter after 10-hour milling H_C=549,1A/m. Further increase in the milling time causes only slight changes of the H_C with slight increasing tendency (Fig. 3).

The longer the milling process, the smaller the value of the saturation of magnetization, which for the powder obtained after 5-hour milling of the amorphous $Co_{77}Si_{11,5}B_{11,5}$ ribbon amounts to B_S=0,85 T. For the powder obtained in 10-hour milling, the value B_S equals 0,858 T. The value of the saturation of induction maintains at the same level for all samples after 15, 20 and 25 hours of milling (B_S=0,86÷0,88 T).

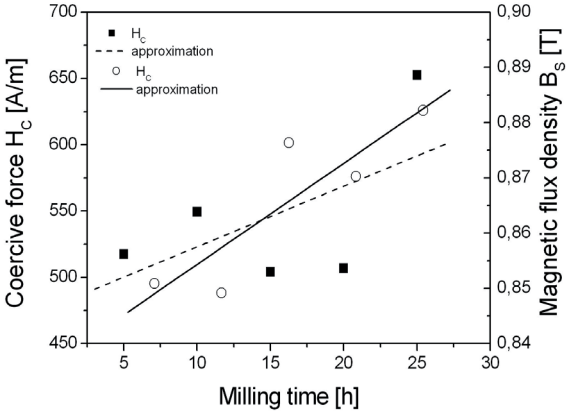

Fig. 3. Coercion field and saturation induction versus milling time of powders obtained from the $Co_{77}Si_{11,5}B_{11,5}$ amorphous ribbons

Magnetic properties of powder materials may be improved by heat treatment. The research revealed that the isothermal annealing causes lowering the coercion field value of the powder material with the saturation induction unchanged. However, the excessively high isothermal annealing temperature causes the significant growth of the coercion field value (Fig. 4A.).

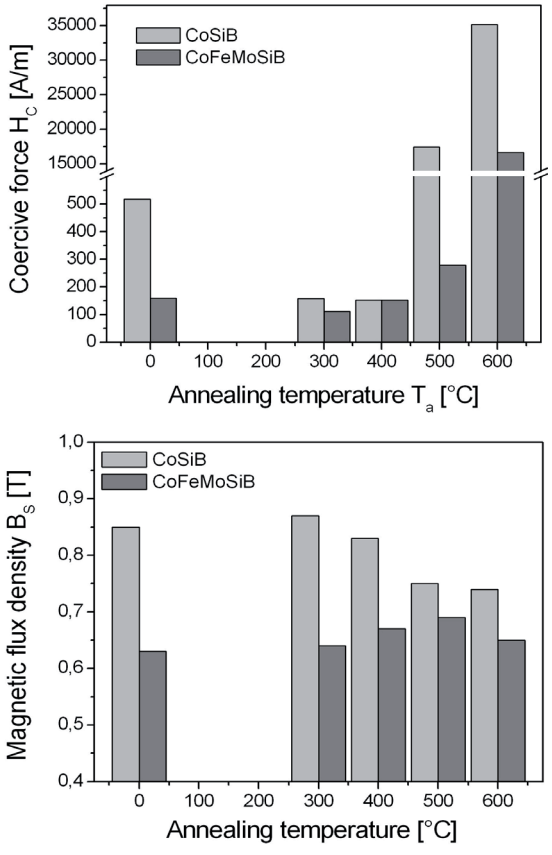

Fig. 4. A) Coercive force H_C, B) Magnetic flux density B_S values versus annealing temperature T_a of the powder obtained from the $Co_{68}Fe_4Mo_1Si_{13,5}B_{13,5}$ and $Co_{77}Si_{11,5}B_{11,5}$ ribbons after 5 hours of high-energy ball milling

The optimum isothermal annealing temperature for the powder material obtained by the high energy milling of the $Co_{68}Fe_4Mo_1Si_{13,5}B_{13,5}$ amorphous ribbon for 5 hours, after which it demonstrates its best soft magnetic properties, is 300°C K for 1 hour (H_C=111.7 A/m). After holding the powder at a temperature of 400°C for 1 hour, its coercion field value increases reaching H_C=152.2 A/m, and the saturation induction reaches B_S=0.67 T. Further powder annealing temperature rise to 500°C causes further growth of the coercion field and saturation induction, which reach values of H_C=279.7 A/m and B_S=0.69 T respectively. However, the coercion field is H_C=16624 A/m and the saturation induction decreases and is B_S=0.65 T after annealing the powder at 600°C K for 1 hour (Fig. 4B).

For powders obtained after 5 hours of milling and at the following stage – heated, similar effects as in the case of amorphous ribbons heating were achieved. Similar results were obtained in the research [14] where the Co-based amorphous alloy ribbons were needed. In this case, the relation of the coercive field to the temperature of heating was similar to the one presented in Fig. 4. It is probable that the isothermal heating of powders provoked the relaxation of stress appeared in the process of high energetic milling. All this is responsible for the change of the magnetic properties of powders.

On the basis of the analysis of the electron diffraction pattern (fig. 5) it may be supposed that apart from the stress relaxation, the process of heating results in the structural changes which consists in new phase nucleation in higher temperatures.

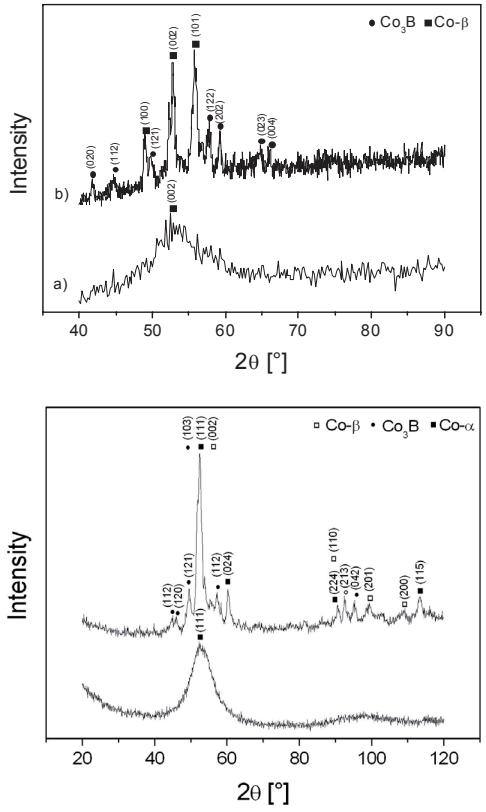

Fig. 5. X-ray phase diagram of the powder A) $Co_{77}Si_{11,5}B_{11,5}$, and B) $Co_{68}Fe_4Mo_1Si_{13,5}B_{13,5}$ obtained after milling for 5 hours and subsequently heated in the temperatures of a) 300 and b) 600°C

It was found out, basing on examinations of thin foils under the transmission electron microscope that the powder obtained after 5 hours of the high energy ball milling and subsequently heated in termal in temperature 600°C/1h in argon has a nanocrystalline structure (Fig. 6). In addition, spectral line from the (111), (002), (113) and Co-α, (011), (022), (023), (121), (024) and Co-β, as well as spectral line from the (021), (103), (222) and Co_3B cobalt boron, were identified in the amorphous structure of the powder.

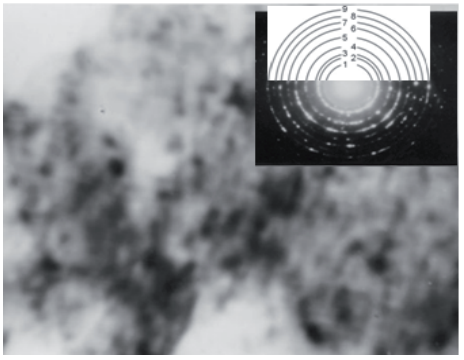

Fig. 6. Structure of the powder grain obtained after 5 hours high energy ball milling amorphous ribbon $Co_{68}Fe_4Mo_1Si_{13,5}B_{13,5}$ and subsequently heated in thermal in temperature 600°C/1h in argon; transmission electron microscope,180000×

4. Conclusions

It was found out in observations on the scanning electron microscope that along with the milling time increase the powder particles size decreases, and that their shape changes also during the process. The powder grains were flake-sized at the first stage of the process, and actually they were parts of the ribbons. However, as the milling time grows the grains become spherical with a clear tendency to get smaller.

The magnetic tests of the $Co_{68}Fe_4Mo_1Si_{13,5}B_{13,5}$ and $Co_{77}Si_{11,5}B_{11,5}$ powders obtained in the high-energy ball of milling process proved that the process causes significant decrease in the magnetic properties. A specially significant is the growing value of coercion field with milling time. The saturation magnetization does not change, or its changes do not have a clear effect on the magnetic properties. As the milling time passes the coercion field decreases; anyway, it grows again with the extended milling time.

On the basis of the research done, it was stated that the process of the high-energy ball milling combined with thermal crystallisation of the $Co_{77}Si_{11,5}B_{11,5}$ and $Co_{68}Fe_4Mo_1Si_{13,5}B_{13,5}$ alloy, results in the production of the nanocrystalline powder material. The structure and magnetic properties of this material may be improved by means of a proper choice of parameters of this process as well as the final thermal treatment.

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