

## Structure and magnetic properties of powder soft magnetic materials

J. Konieczny <sup>a</sup>, L.A. Dobrzański <sup>a,\*</sup>, A. Przybył <sup>b</sup>, J.J. Wysocki <sup>b</sup>

<sup>a</sup> Division of Materials Processing Technology, Management and Computer Techniques in Materials Science, Institute of Engineering Materials and Biomaterials, Silesian University of Technology, ul. Konarskiego 18a, 44-100 Gliwice, Poland

<sup>b</sup> Faculty of Materials Processing Technology and Applied Physics, Częstochowa University of Technology, ul. Armii Krajowej 19, 42-200 Częstochowa, Poland

\* Corresponding author: E-mail address: leszek.dobrzanski@polsl.pl

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### Materials

#### ABSTRACT

**Purpose:** The paper presents influence of high-energy mechanical milling process, isothermal annealing and to a combination of these two technologies of cobalt base metallic glasses  $\text{Co}_{77}\text{Si}_{11.5}\text{B}_{11.5}$  on magnetic properties and their structure.

**Design/methodology/approach:** The powder test piece obtained from the input amorphous ribbon in high-energy ball milling. The diffraction examinations and examinations of thin foils were made on the JEOL JEM 200CX transmission electron microscope. Observations of the structure of powders were made on the Opton DSM-940 scanning electron microscope. The X-ray tests were realized with the use of the XRD 7 SEIFERT-FPM diffractometer.

**Findings:** analysis of the magnetic properties test results of the of the  $\text{Co}_{77}\text{Si}_{11.5}\text{B}_{11.5}$  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 metal powders 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 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 as well as structure and magnetic properties of investigated samples is presented.

**Keywords:** Nanomaterials; Powders; Magnetic properties; Composites

### 1. Introduction

Research on nanocrystalline materials obtained from metallic glasses crystallisation has been started in 1988 by Yoshizava [1]. Since then those materials have been intensively studied mostly

because of their excellent soft magnetic properties [2-4]. The nanocrystallisation process can be realised in several ways. Thermal nanocrystallisation or mechanical milling of metallic glasses agged the most frequently [5-9].

Because of the manufacturing process the metallic nanocrystalline materials obtained directly from the metallic

glasses crystallisation are available only in the form of super thin strips. Nanocrystalline materials in a powder state (loose) seem to be a very interesting problem from a production technology, processing and application points of view [10, 11]. The obtention of powder nanocrystalline materials in the high energy ball mill allows to carry out research on ferromagnetic nanocomposites whose shape and dimensions can be freely formed with the use of different consolidation techniques [12-16].

## 2. Experimental

The investigations were carried out on a  $\text{Co}_{77}\text{Si}_{11,5}\text{B}_{11,5}$  metallic glass in form of 0,036 mm thick and 10,2 mm wide strips. A 8000 SPEX CertiPrep Mixer/Mill high energy ball mill was applied to mill the strips both in „as quenched” state and heat treated. The vibration times were 5, 10, 15, 20, 25, 30 and 60 hours. A THERMOLYNE F6020C resistance furnace was used for isothermal soaking of the amorphous strips and the powder. Soaking was carried out at temperature range 300-600°C with 100°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  $2\theta$  angle range from 40 to 120° (measurement step 0,1°). Pulse counting time was 5 s.

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

To measure the grain size of the powder a LEICA light microscope and appropriate computer programme was applied. 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  $\text{Co}_{77}\text{Si}_{11,5}\text{B}_{11,5}$  strip lasting 5 hours, results in the production of flakes of strips (“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). The microscope observation proves that the longer the process of milling the smaller the value of the average diameter of powder grains and the smaller the value of the standard deviation of the average diameter of grains.

At the first stage of the milling process the powder obtained was in the form of flakes of the crumbled string of the average dimension 230,9  $\mu\text{m}$  (standard deviation  $s=163,9$  5-hour milling). After 10-hour milling, the average diameter of powder grains decreases to 89,7  $\mu\text{m}$  (standard deviation  $s=59,5$ ). At the second stage of the process, the time of milling still rises and the average diameter of powder grains still decreases but with smaller intensity. At the final stage of the tests, after 60-hour high energy ball milling, the powder grains are characterised by the equiaxial shape and the distinct size diversity (Fig. 2).

The analysis of powder grain dimensions proved that the value of the average of diameter of powder grains decreases along with the milling time increase. The research proves that there is a process of size homogenisation, even though the range of size of powder grains is still significant.

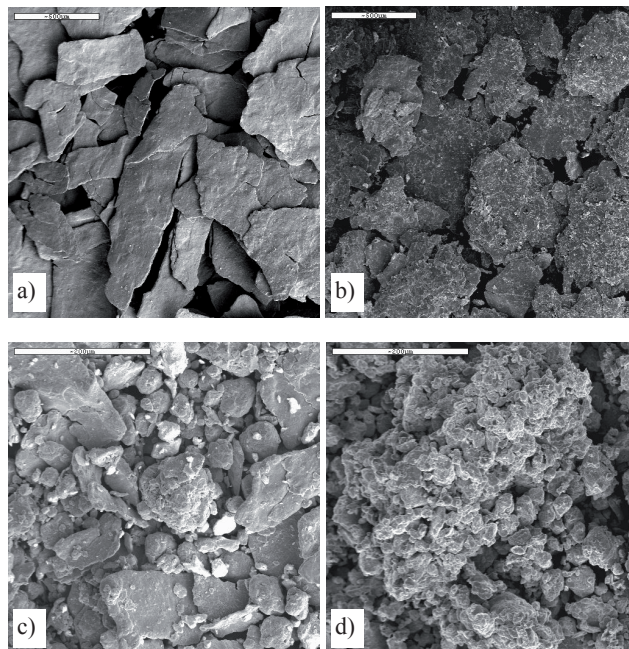


Fig. 1. Powder grains image after a) 5 h, b) 10 h, c) 20, d) 30 h of the high energy ball milling, scanning microscope

It was found out, basing on examinations of thin foils under the transmission electron microscope that the powder obtained after 25 and 60 hours of the high energy ball milling of the amorphous strip has a nanocrystalline structure (Fig. 3). In addition, spectral line from the (101), (103) and  $\text{Co-}\beta$ , as well as spectral line from the (022), (212) and  $\text{Co}_3\text{B}$  cobalt boron, were identified in the amorphous structure of the powder.

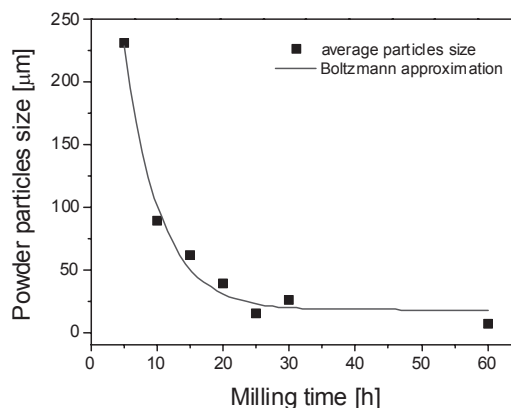


Fig. 2. Effect of the milling time for the amorphous strips made out of  $\text{Co}_{77}\text{Si}_{11,5}\text{B}_{11,5}$  alloy on the powder particles size for the obtained powders

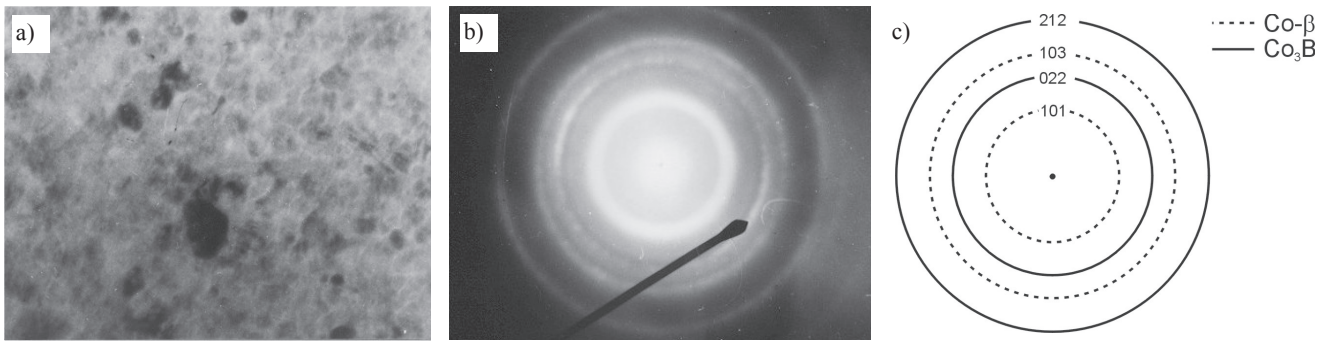


Fig. 3. a) Structure of the powder grain obtained after 25 hours of the high energy ball milling of the amorphous strip from the  $\text{Co}_{77}\text{Si}_{11,5}\text{B}_{11,5}$  alloy; transmission electron microscope, b) electron diffraction pattern from the structure are shown in Fig 3a), c) solution of the diffraction pattern from Fig. 3b

The magnetic research of the  $\text{Co}_{77}\text{Si}_{11,5}\text{B}_{11,5}$  powders obtained in the process of milling of the strips 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 strip 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  $H_C=549,1\text{A/m}$ . Further increase in the milling time causes only slight changes of the  $H_C$  with slight increasing tendency (Fig. 4).

The longer the milling process, the smaller the value of the saturation of magnetisation, which for the powder obtained after 5-hour milling of the amorphous  $\text{Co}_{77}\text{Si}_{11,5}\text{B}_{11,5}$  strip amounts to  $B_S=0,85\text{ T}$ . For the powder obtained in 10-hour milling, the value  $B_S$  equals  $0,858\text{ T}$ . The value of the saturation of induction maintains at the same level for all samples after 15, 20, 25 and 30 hours of milling ( $B_S=0,86\pm 0,88\text{ T}$ ).

At the next stage of the experiment, the powder material was subjected to the isothermal heating in order to determine the influence of the temperature on the change of the magnetic properties. Initial powders were obtained as the result of 5 hour high energy ball milling of the amorphous  $\text{Co}_{77}\text{Si}_{11,5}\text{B}_{11,5}$  alloy.

The lowest value of the coercive force  $H_C=152\text{ A/m}$  belongs to the powder obtained in the amorphous strip milling lasting for 5 hours and then heated in the temperature of  $400^\circ\text{C}$  for an hour. The magnetic properties of the powder material heated in the definite temperatures are presented in Fig 5.

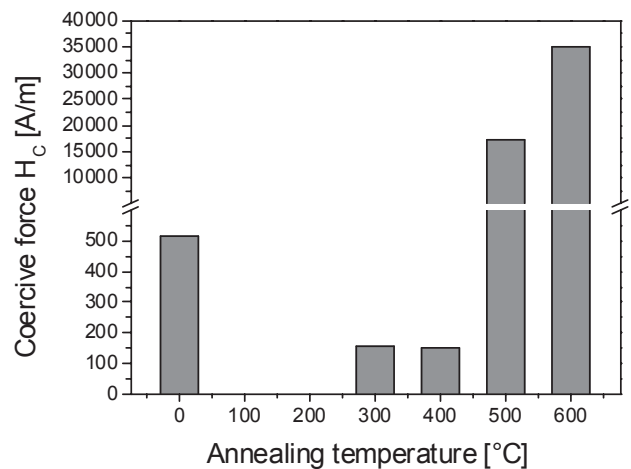


Fig. 5. Effect of the isothermal heating of the powder on the coercive field intensity  $H_C$  and saturation induction  $B_S$ . The powder was obtained by the high-energy milling the  $\text{Co}_{77}\text{Si}_{11,5}\text{B}_{11,5}$  amorphous alloy for 5 hours

For powders obtained after 5 hours of milling and at the following stage – heated, similar effects as in the case of amorphous strips heating were achieved. Similar results were obtained in the research [16] 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. 5. 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. 6) it may be supposed that apart from the stress

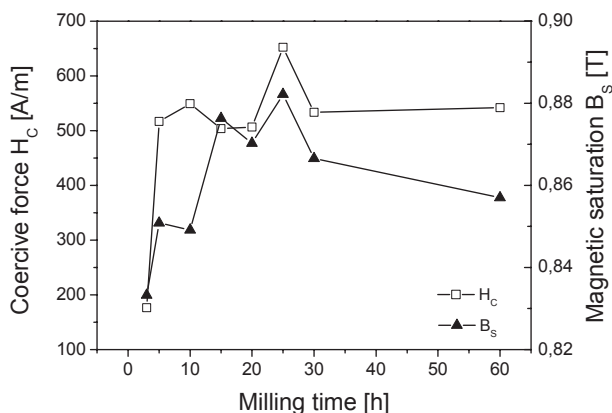


Fig. 4. Effect of the milling time for the amoprhus ribbons made from the  $\text{Co}_{77}\text{Si}_{11,5}\text{B}_{11,5}$  alloy on the coercive force intensity  $H_C$  and magnetisation  $B_S$  for the obtained powders

relaxation, the process of heating results in the structural changes which consists in new phase nucleation in higher temperatures.

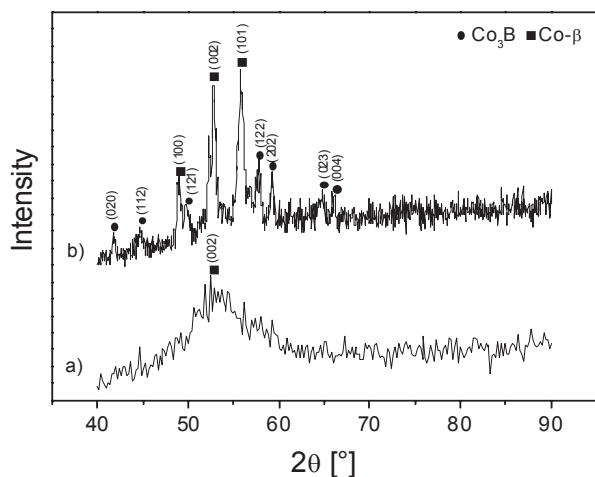


Fig. 6. X-ray phase diagram of the powder material obtained after milling for 5 hours and subsequently heated in the temperatures of a) 300 and b) 600°C

## 4. Conclusions

The magnetic tests of the  $\text{Co}_{77}\text{Si}_{11.5}\text{B}_{11.5}$  powders obtained in the high-energy ball of milling process proved that the process causes significant decrease in the magnetic properties.

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  $\text{Co}_{77}\text{Si}_{11.5}\text{B}_{11.5}$  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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