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# Microstructure investigations of Co-Si-B alloy after milling and annealing

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**Methodology of research**

## ABSTRACT

**Purpose:** The work presents the microstructure characterization of $Co_{77}Si_{11,5}B_{11,5}$ metallic glass after high-energy ball milling and heat treatment processes.
**Design/methodology/approach:** The studies were performed on ribbon prepared by melt spinning and this ground in high-energy vibratory ball mill. The tested ribbon and obtained powders were also annealed in specified heat treatment conditions. The morphology of the powder particles of milled ribbon was analyzed by using the confocal laser scanning microscope. The methods of X-ray diffraction were used for the qualitative phase analysis. The parameters of the individual diffraction line profiles were determined by PRO-FIT Toraya procedure. The average crystallite sizes and lattice distortions for Co phase were estimated using Williamson-Hall method.
**Findings:** The studied $Co_{77}Si_{11.5}B_{11.5}$ metallic glass in annealed state contains hexagonal Co crystalline phases emerged in amorphous matrix. The crystallite size of Co phase in as-cast sample lies in nanoscale. After annealing process the crystallite size increases to 72 nm and diminishes to 46 nm in the powder sample after 30 hours of milling. The milling causes decrease of the crystallite size and increase of lattice distortions of crystalline phase. The powder particles after 30 hours of milling are of spherical shape.
**Practical implications:** The powder particles obtained after milling process of Co-based metallic glass could be suitable components in production of ferromagnetic nanocomposites.
**Originality/value:** The obtained results confirm the utility of applied investigation methods in the microstructure analysis of powder materials with nanocrystalline phases.
**Keywords:** X-ray phase analysis; Toraya procedure; High-energy ball milling; Metallic glasses

## 1. Introduction

The metallic glasses are the most interesting materials for technological applications due to their special and unique mechanical, magnetic and chemical properties. The formation of the amorphous state depends on the alloy composition and manufacturing process conditions. The rapid solidification of liquid metallic melts [1-6] is the most often used for metallic glass preparation. These materials can be also manufactured by electrodeposition and sputtering, which allow producing the samples of thin film, ribbon, wire, tube and also bulk forms [7-9].

The metallic glasses are suitable materials for many applications such as electronic measuring and surveillance systems, magnetic wires, sensors, band-pass filters, magnetic shielding, energy-saving electric power transformers [10,11].

High-energy ball milling can result in the mechanical alloying, when it involves a mixture of powders and also in

conventional milling, when it involves a pure elements or mixtures [12].

High-energy ball milling process leads to a generation of a large amount of lattice defects, distortions and deformations, which enable the structure modification of milled material. During high-energy ball milling the powder particles undergo different forms of ball impacts, which induce mechanical deformation, cold welding and fracture processes [13].

High-energy ball milling is an effective method to prepare compounds, composites, amorphous phases, nanocrystalline materials and also superconducting materials, rare permanent magnets, superplastic alloys [14].

## 2. Material and research methodology

The aim of the present work was the microstructure characterization of $Co_{77}Si_{11.5}B_{11.5}$ metallic glass using XRD (X-Ray Diffraction) and CLSM (Confocal Laser Scanning Microscopy) methods. The studies were performed on ribbon prepared by melt spinning and this ground in high-energy vibratory ball mill. The tested ribbon and obtained powders were also annealed in different heat treatment conditions.

The X-ray diffraction and electron microscopy methods are of great importance in the microstructure characterization of complex, multiphase materials. The application of X-ray diffraction methods enables not only qualitative and quantitative phase analysis, but also microstructure characterization (crystallite size, lattice distortions, dislocation densities, stacking faults and twins probability).

The values of *FWHM* (Full Width at Half Maximum) parameters of diffraction lines were determined using Toraya PRO-FIT procedure [15]. The average crystallite sizes and lattice distortions for Co phase were estimated using Williamson-Hall method. The main assumption of the Williamson-Hall method is that the physical line broadening *FWHM* is a sum of line broadening connected to crystallite size ($\beta_D$) and of line broadening connected to lattice strain ($\beta_{LS}$) following to an equation:

$$FWHM = \beta_D + \beta_{LS} \tag{1}$$

The physical line broadening is obtained from relation:

$$\beta = FWHM - \beta_S \tag{2}$$

where:
$\beta$ – physical broadening (from the sample),
$\beta_S$ – instrumental broadening (from the standard).

The NIST SRM660a ($LaB_6$ powder) was used as a line profile standard to determine the instrumental broadening.

The ball milling process was realized on $Co_{77}Si_{11,5}B_{11,5}$ metallic glass. The studied material was cast as metallic glass in the form of ribbon with thickness 0.03 mm and width 9.3 mm. The ribbons were manufactured by the melt spinning technique, which is a method of continuous casting of the liquid alloy on surface of turning copper roller.

The chemical composition and dimensions of ribbon of examined alloy are presented in Table 1.

Table 1
Chemical composition and dimensions of ribbon of investigated metallic glass

| Chemical constitution | Co | Si | B | Dimensions of ribbon | Thickness | Width |
|---|---|---|---|---|---|---|
| at.% | 77 | 11.5 | 11.5 | mm | 0.03 | 9.3 |

Ribbons of the tested matallic glass were cut into pieces about 10 mm x 9 mm. The high-energy ball milling process was realized in a vibratory mill type SPEX 8000 CertiPrep Mixer/Mill (Fig. 1) for 10, 20 and 30 hours under argon atmosphere.

The mill was equipped with a stainless container (diameter Ø = 38 mm, height h = 63 mm) and balls (four steel balls – diameter Ø = 6.3 mm and two steel balls – diameter Ø = 12.7 mm). The weight ratio of balls to milled material was 2.5:1. The mill generated vibrations of the balls and a material inside the container during which their collisions occur.

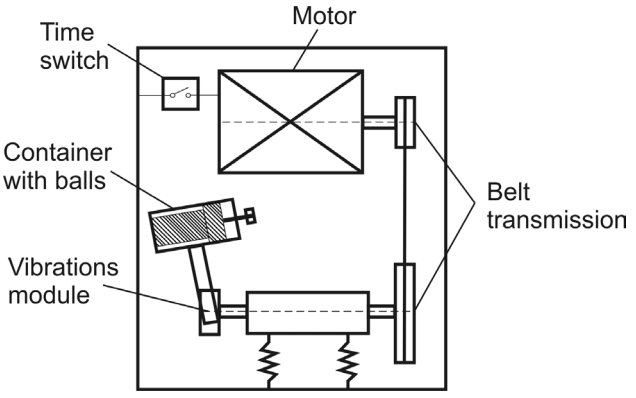


Fig. 1. Schematic illustration of SPEX 8000 CertiPrep Mixer/Mill construction

Powders and ribbons of studied metallic glass were annealed in electric chamber furnace THERMOLYNE 6020C under protective argon atmosphere at temperature of 823 K. The annealing time was constant and equal to 1 hour.

Phase analysis was carried out using the X-Pert Philips diffractometer equipped with curved graphite monochromator on diffracted beam and a tube provided with copper anode. It was supplied by current intensity of 30 mA and voltage of 40 kV. The length of radiation ($\lambda Cu_{K\alpha}$) was 1.54178 Å. The data of diffraction lines were recorded by "step-scanning" method in $2\theta$ range from 20° to 100° and 0.05° step.

The morphology of the powder particles of milled ribbon was analyzed by using of OLYMPUS LEXT OLS3000 confocal laser scanning microscope. The observations were made in real time under atmospheric conditions.

## 3. Results and discussion

The X-ray diffraction investigations revealed that the studied $Co_{77}Si_{11.5}B_{11.5}$ metallic glass in as-cast state contains crystalline phase emerged in amorphous matrix (broad diffraction halo

characteristic for the amorphous structure). The diffraction pattern of this sample after annealing at 823 K for 1 hour shows the single peaks from (100), (002), (101), (110) and (112) planes of hexagonal Co ($P6_3/mmc$) phase (Fig. 2).

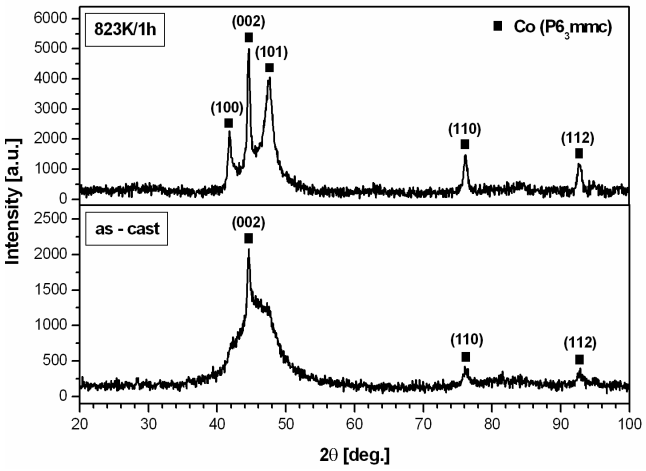


Fig. 2. X-ray diffraction pattern of $Co_{77}Si_{11,5}B_{11,5}$ ribbon in as-cast state and annealed at 823 K for 1 hour

Diffraction patterns of the samples after high-energy ball milling process and heat treatment (Fig. 3) show the broadening of diffraction lines. These effects indicate that high-energy ball milling causes decrease of the crystallite size of crystalline phase and increase of its lattice distortions. Thus the main result of milling process of $Co_{77}Si_{11.5}B_{11.5}$ ribbon is the increasing content of the amorphous phase.

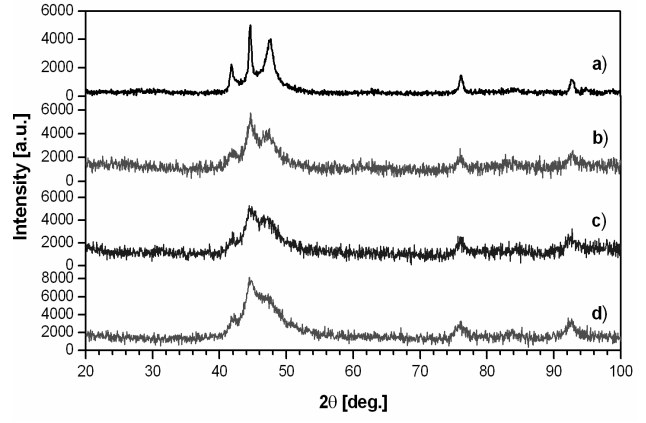


Fig. 3 X-ray diffraction patterns of $Co_{77}Si_{11,5}B_{11,5}$ alloy after annealing at 823 K and different times of high-energy ball milling: a) 0 h, b) 10 h, c) 20 h, d) 30 h

Figure 4 shows the morphology of powder particles of the samples as a result of ribbon milling observed by the confocal laser scanning microscope. After 30 hours of high-energy ball milling process the shape of powder particles is spherical. Moreover, the powder particles are irregular.

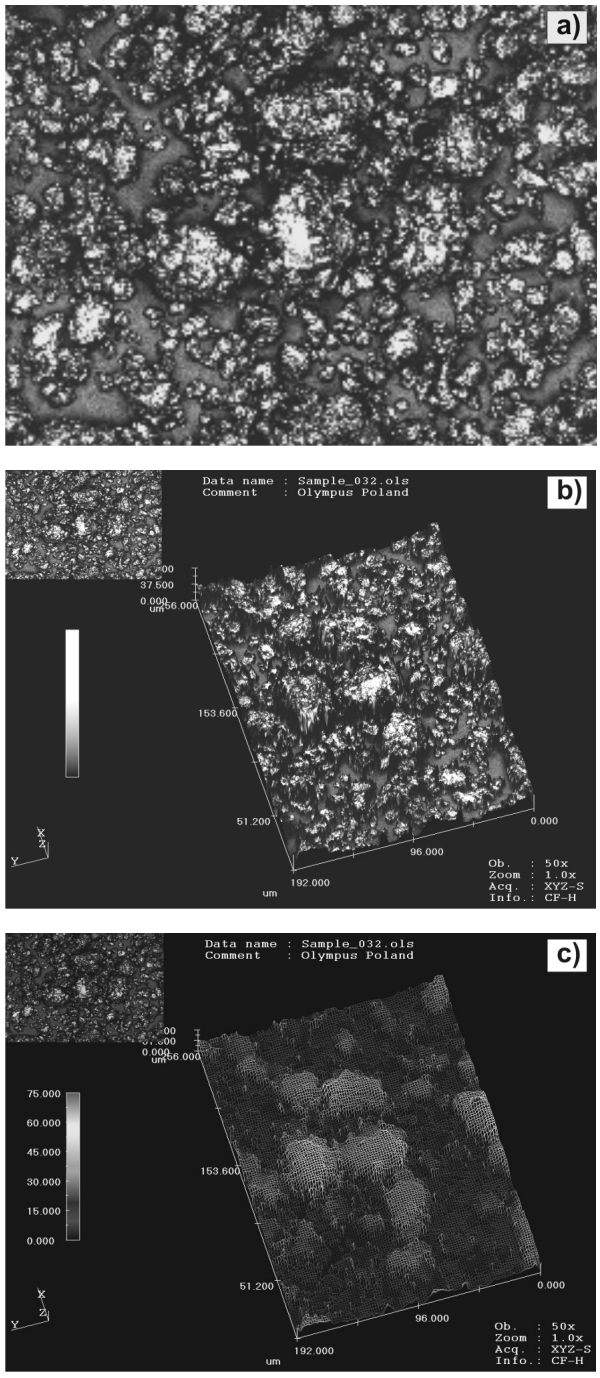


Fig. 4. Morphology of powder particles of $Co_{77}Si_{11,5}B_{11,5}$ metallic glass after 30 hours of milling process: a) conventional 2D profile, b,c) confocal 3D profiles

The determination of *FWHM* parameters of individual diffraction line profiles was performed by PRO-FIT Toraya's procedure. The changes of an averaged crystallite size ($D$) and of the lattice distortion ($<\Delta a/a>$) with the increase of the high-energy ball milling time for Co phase are presented in Fig. 5.

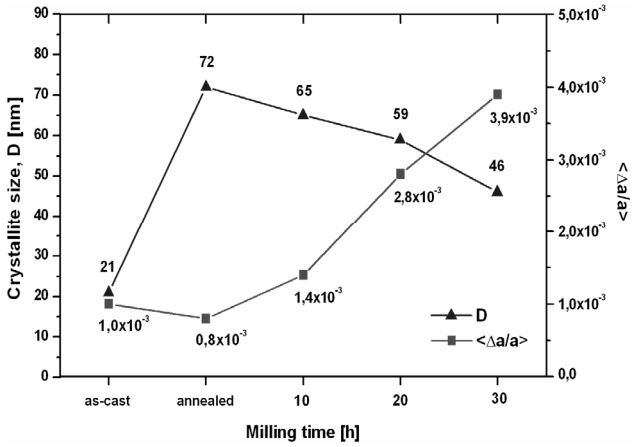


Fig. 5 Changes of averaged crystallite size ($D$) and lattice distortion ($<\Delta a/a>$) with the increase of milling time for Co phase

The crystallite size ($D$) of Co phase in as-cast sample lies in nanoscale and is equal to 21 nm. After annealing process the crystallite size increases to 72 nm and diminishes to 46 nm in the powders sample after 30 hours of milling (Fig. 5).

The increase of lattice distortion ($<\Delta a/a>$) of Co phase with the increase of milling time is also studied (Fig. 5). The minimum of lattice distortion is observed for ribbon sample after heat treatment, but before milling process.

## 4. Conclusions

The investigations performed on the ribbon of $Co_{77}Si_{11.5}B_{11.5}$ metallic glass and this ground in high-energy vibratory mill allowed to formulate the following statements:

- in annealed at 823 K ribbon Co crystalline phase embedded in amorphous matrix was found,
- the crystallite size of Co phase in as-cast, annealed and ground states lies in nanoscale,
- the high-energy ball milling process causes decrease of the crystallite size of Co phase and increase of lattice distortions,
- the powder particles after 30 hours of milling are irregular and have spherical shape.

## References

[1] R. Nowosielski, R. Babilas, Fabrication of bulk metallic glasses by centrifugal casting method, Journal of Achievements in Materials and Manufacturing Engineering 20 (2007) 487-490.

[2] R. Nowosielski, S. Griner, Shielding of electromagnetic fields by mono- and multi-layer fabrics made of metallic glasses with Fe and Co matrix, Journal of Achievements in Materials and Manufacturing Engineering 20 (2007) 45-54.

[3] R. Nowosielski, L.A. Dobrzański, J. Konieczny, Influence of temperature on structure and magnetic properties of powders alloys, Journal of Achievements in Materials and Manufacturing Engineering 20 (2007) 163-166.

[4] J. Konieczny, L.A. Dobrzański, A. Przybył, J.J. Wysłocki, Structure and magnetic properties of powder soft magnetic materials, Journal of Achievements in Materials and Manufacturing Engineering 20 (2007) 139-142.

[5] D. Szewieczek, T. Raszka, J. Olszewski, Optimisation the magnetic properties of the $(Fe_{1-x}Co_x)_{73.5}Cu_1Nb_3Si_{13.5}B_9$ (x=10; 30; 40) alloys, Journal of Achievements in Materials and Manufacturing Engineering 20 (2007) 31-36.

[6] H. Chiriac, T.A. Ovari, Amorphous glass-covered magnetic wires: preparation, properties, applications, Progress in Materials Science 40 (1996) 333-407.

[7] J.M. Garcia, A. Asenjo, D. Garcia, C. Prados, M. Vazquez, Properties of amorphous magnetic materials by magnetic force microscopy, Journal of Non-Crystalline Solids 287 (2001) 55-59.

[8] P. Gramatyka P., A. Kolano-Burian, R. Kolano, M. Polak, Nanocrystalline iron based powder cores for high frequency applications, Journal of Achievements in Materials and Manufacturing Engineering 18 (2006) 99-102.

[9] S. Lesz, D. Szewieczek, J.E. Frąckowiak, Structure and magnetic properties of amorphous and nanocrystalline $Fe_{85.4}Hf_{1.4}B_{13.2}$ alloy, Journal of Achievements in Materials and Manufacturing Engineering 19 (2006) 29-34.

[10] B. Ziębowicz, D. Szewieczek, L.A. Dobrzański, New possibilities of application of composite materials with soft magnetic properties, Journal of Achievements in Materials and Manufacturing Engineering 20 (2007) 207-210.

[11] P. Vojtanik, Magnetic relaxations in amorphous soft magnetic alloys, Journal of Magnetism and Magnetic Materials 304 (2006) 159-163.

[12] D.L. Zhang, Processing of advanced materials using high-energy mechanical milling, Progress in Materials Science 49 (2004) 537-560.

[13] M. Pękała, M. Jachimowicz, V.I. Fadeeva, H. Matyja, Phase transformations in Co-B-Si alloys induced by high-energy ball milling, Journal of Non-Crystalline Solids 287 (2001) 360-365.

[14] C. Zhenhua, C. Ding, C. Gang, Y. Hongge, H. Peiyun, Preparation of elevated-temperature intermetallic powders via a novel reaction ball milling technique, Journal of Alloys and Compounds 370 (2004) 43-46.

[15] G.K. Williamson, X-ray line broadening from filed aluminium and wolfram, Acta Metallurgica 1 (1953) 22-31.