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The production attempt of Fe-Nb-B-Zr and Fe-Nb-B-Y system alloys by die pressure casting method

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Materials

ABSTRACT

Purpose: The purpose of this article was to investigate the possibility of the production of Fe-Nb-B bulk metallic alloys with additions of yttrium and zirconium elements. Furthermore, this paper tends to present the structure and selected properties of obtained alloys. In this article the influence of an argon atmosphere on casting process was observed too.

Design/methodology/approach: The production attempts were performed on Fe-Nb-B-Zr and Fe-Nb-B-Y system alloys in form of a plate. Master alloy ingots with compositions of $Fe_{72}B_{22}Y_4Nb_2$ and $Fe_{71}(Nb_{0.8}Zr_{0.2})_6B_{23}$ were prepared by induction melting of pure Fe, Nb, B, Y and Fe, Nb, B, Zr elements in an argon atmosphere. The ingots have been crushed and then the investigated material was cast with and without a protective atmosphere. The investigated materials were cast in form of a plate with thickness of 1 mm. The structure analysis of the studied materials in as-cast state was carried out using X-ray diffraction (XRD) and microscopic observation. The thermal properties of the alloys were examined by DSC methods. The measurments of the hardness were performed with the Vickers method.

Findings: The Fe-Nb-B-Y and Fe-Nb-B-Zr system alloys in form of a plate were produced by die pressure casting method. The investigation methods revealed that the studied as-cast alloys were crystalline. The structure of the obtained plates is rather fine-grained and there were not found any impurities and undesirable phases inside the materials. The results of calorimetric curves confirm that all tested samples are crystalline.

Practical implications: To extend the potential applications of the Fe-based BMGs, amorphous alloys with larger critical sizes and better processability are required. The Fe-Nb-B-Zr and Fe-Nb-B-Y bulk metallic glasses obtained by die pressure casting method can be used for production of telecommunications devices, sensors or low-energy transformers. These materials exhibit excellent mechanical and soft magnetic properties.

Originality/value: An overall presentation of an influence of the yttrium and zirconium additions on the attempt of forming $Fe_{72}B_{22}Y_4Nb_2$ and $Fe_{71}(Nb_{0.8}Zr_{0.2})_6B_{23}$ alloys. The chemical composition of these alloys was tested in our laboratory for the first time.

Keywords: Amorphous materials; Bulk metallic glasses; Fe-based alloy; Hardness; Microscopic observation

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1. Introduction

In the last years, the Fe-based bulk metallic glasses (BMG) have attracted attention because of their good glass forming ability and excellent mechanical properties [1-3]. One of the most important and well recognized amorphous alloys is Fe-Co-B-Si-Nb system alloy, because of excellent magnetic properties, the absence of special toxic elements and good glass forming ability.

Recently, the development of Fe-based BMGs with higher GFA in conjunction with high strength has been required [4-6]. From the analysis of literature data [7-11] it can be concluded that yttrium or zirconium elements, even in small addition have been shown to be advantageous in bulk glass formation. The proper addition of these elements should have the influence on the glass forming ability and improved brittleness of the bulk metallic glasses-forming alloys. Therefore, there is the need for more investigations in this field.

The aim of this work was an attempt to produce Fe-Nb-B bulk metallic alloys with additions of yttrium and zirconium elements. The additional objectives were presentation of the structure and selected properties of the obtained alloys and observation of the influence of an argon atmosphere on casting process.

2. Materials for research and work methodology

Fe-based master alloy ingots with compositions (Table 1-3) of $Fe_{72}B_{22}Y_4Nb_2$ and $Fe_{71}(Nb_{0,8}Zr_{0,2})_6B_{23}$ were prepared by induction melting of pure Fe, Nb, B, Y, Zr elements in an argon atmosphere. Applied ingots were melted one at a time. The composition of alloys represents nominal atomic percentages. The ingots have been crushed and then the investigated material was cast by the pressure die casting method in an argon atmosphere and without a protective atmosphere.

Table 1.
Chemical composition of $Fe_{72}B_{22}Y_4Nb_2$ alloy

No	Elements	Mass. [%]	At. [%]
1	Fe	83.76	72
2	Nb	3.87	2
3	B	4.96	22
4	Y	7.41	4

Table 2.
Chemical composition of $Fe_{71}(Nb_{0,8}Zr_{0,2})_6B_{23}$ alloy

No	Elements	Mass. [%]	At. [%]
1	Fe	83.14	71
2	Nb	9.35	4.8
3	B	5.22	23
4	Zr	2.29	1.2

Table 3.
Purity and shape of used elements

No	Elements	Purity [%]	Shape
1	Fe	99.97	pieces
2	Nb	99.8	pieces
3	B	99.5	pieces
4	Y	99.9	lump
5	Zr	99.9	lump

The selected compositions of alloys fulfill specific conditions for iron based bulk metallic glass production:

- at least the alloys are ternary,
- at least two components of these system alloys are metallic (Fe, Zr, Nb, Y),
- metal elements have different radii (Fe - 0.12412 nm [7], Zr - 0.16025 nm [7], Nb - 0.1429 nm [7], Y - 0.181 nm [1, 8]),
- metal and semi-metal pairs have a large negative heat of mixing:
  - Fe-B = - 26 KJ/mol [7, 9],
  - Zr-B = - 56 KJ/mol [10],
  - Y-B = - 35 KJ/mol [11],
  - Nb-B = - 54 KJ/mol [12],
- semi-metals elements participation in the alloy is approximately 20%.

The studies were carried out on bulk metallic alloys in form of plates of 1 mm thickness (Fig. 1).

a) b)

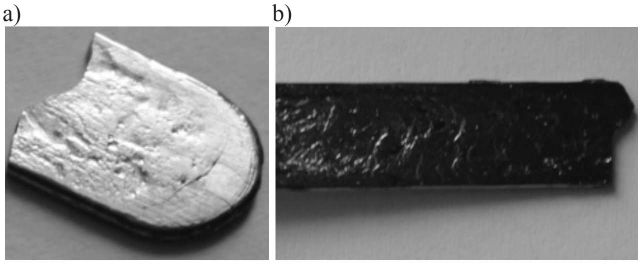

Fig. 1. Images of tested samples of Fe-Nb-B-Y (a) and Fe-Nb-B-Zr (b) system alloys obtained by the pressure die casting method

The structure analysis of the studied materials in as-cast state was carried out using X-ray diffraction and microscopic observation.

Crystalline structures were tested with X-ray diffraction (XRD) using a Seifert - FPM XRD 7 diffractometer with Co Kα radiation at 35 kV. The data of diffraction lines were recorded by means of the stepwise method within the angular range of 20° to 100° and the counting time in the measuring point was 2 s. The identification of tested alloy compounds was carried out using the JCPDS-ICDD catalogs.

The observation of the shape and size of the grains was carried out by means of an OLYMPUS optical microscope GX71 model. The test specimens were mounted, then polished by grinding and polishing machine Tegramin-30 by Struers. Mechanical polishing was performed on a LaboPol Struers-35 mill. The samples were etched. The etching time was 5-10 s.

The thermal properties of the obtained alloys were examined by DSC method. The analysis of the crystallization process was carried out using DSC882e Mettler-Toledo calorimeter at a constant heating rate of 40 K/min. The tests were carried out under an argon atmosphere.

Then, there were performed the measurements of the hardness with the Vickers method. The hardness measurements were carried out by means of Future-Tech FM-700 microhardness tester. The results have been converted from the Vickers scale into the Rockwell scale, too.

3. Results of researches

3.1. X-ray analysis

The bulk metallic alloys samples in form of plates were produced and investigated. The structures of as-cast $Fe_{72}B_{22}Y_4Nb_2$ and $Fe_{71}(Nb_{0,8}Zr_{0,2})_6B_{23}$ alloys in form of plates of 1 mm thickness were examined by X-ray diffraction method.

The diffraction records of sample plates of $Fe_{72}B_{22}Y_4Nb_2$ alloy obtained in an argon atmosphere and without a protective atmosphere are shown in Fig. 2. and Fig. 3. The diffraction patterns recorded for the plates of $Fe_{72}B_{22}Y_4Nb_2$ alloy show the peaks characteristic for Fe_2B, $Fe_{23}B_6$, NbB phases. Whereas, Fig. 4. and Fig. 5. show the X-ray diffraction patterns of the $Fe_{71}(Nb_{0,8}Zr_{0,2})_6B_{23}$ alloy plates of 1 mm thickness obtained in an argon atmosphere and without a protective atmospheres. The diffraction patterns of $Fe_{71}(Nb_{0,8}Zr_{0,2})_6B_{23}$ alloy plates recorded for both samples show the peaks characteristic for Fe_2B, α-Fe and NbB phase.

The X-ray analysis shows that there are no significant differences between the alloy of the same chemical composition obtained in an argon atmosphere and without a protective atmospheres.

The Fe-based samples of determined chemical composition consist of a crystalline phase as was evidenced from crystalline peaks. Investigations did not show the presence of an amorphous structure in the studied samples. Furthermore, X-ray analysis did not reveal the presence of phases with yttrium and zirconium elements. Probably, the yttrium and zirconium atoms form the solid solutions with other metals. There is a possibility of appearance of yttrium and zirconium concentrations, too.

3.2. Thermal analysis

The thermal stability of the as-cast $Fe_{72}B_{22}Y_4Nb_2$ and $Fe_{71}(Nb_{0,8}Zr_{0,2})_6B_{23}$ alloys were studied. The main objective of this analysis was to determine the glass transition temperature and crystallization temperature.

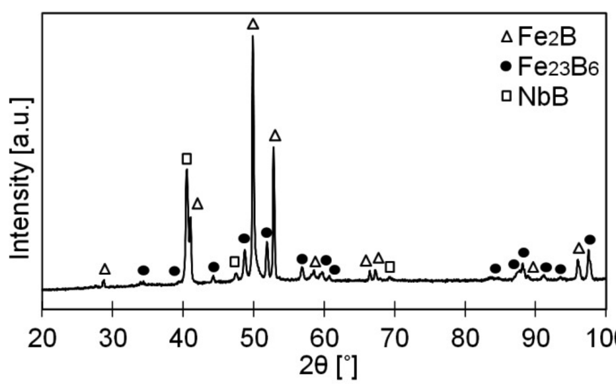

Fig. 2. X-ray diffraction pattern of as-cast $Fe_{72}B_{22}Y_4Nb_2$ alloy plate of 1 mm thickness obtained without a protective atmosphere

The DSC curves measured at 40 K/min. on the plates of $Fe_{72}B_{22}Y_4Nb_2$ and $Fe_{71}(Nb_{0,8}Zr_{0,2})_6B_{23}$ alloys of 1 mm thickness in as-cast state are shown in Fig. 6.

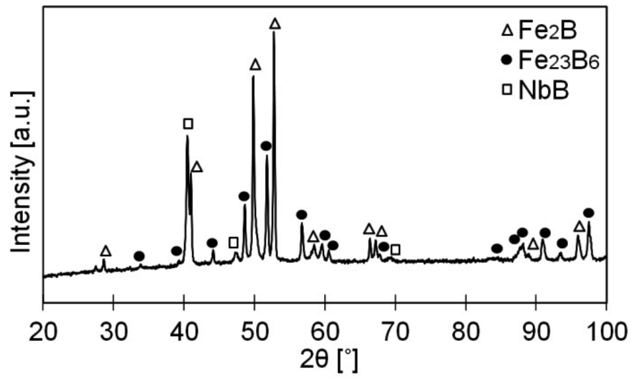

Fig. 3. X-ray diffraction pattern of as-cast $Fe_{72}B_{22}Y_4Nb_2$ alloy plate of 1 mm thickness obtained with a protective atmosphere

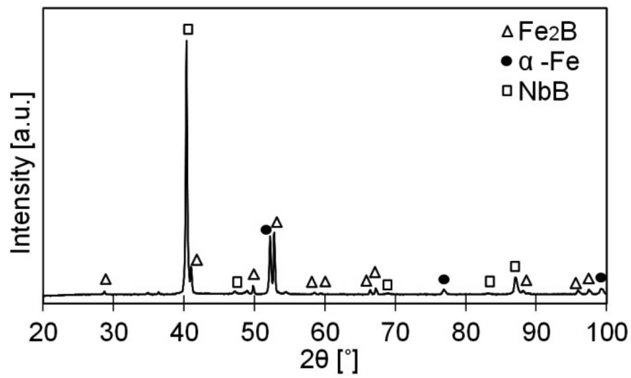

Fig. 4. X-ray diffraction pattern of as-cast $Fe_{71}(Nb_{0,8}Zr_{0,2})_6B_{23}$ alloy plate of 1 mm thickness obtained without a protective atmosphere

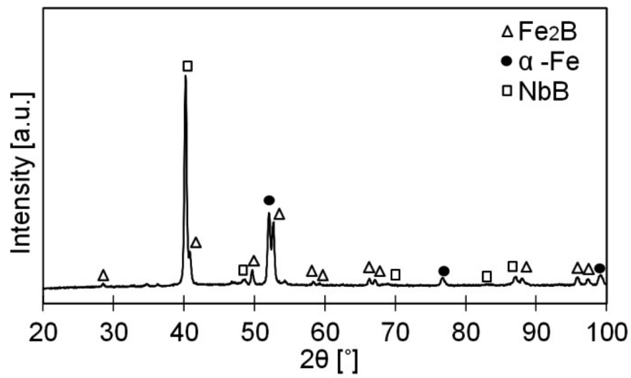

Fig. 5. X-ray diffraction pattern of as-cast $Fe_{71}(Nb_{0,8}Zr_{0,2})_6B_{23}$ alloy plate of 1 mm thickness obtained with a protective atmosphere

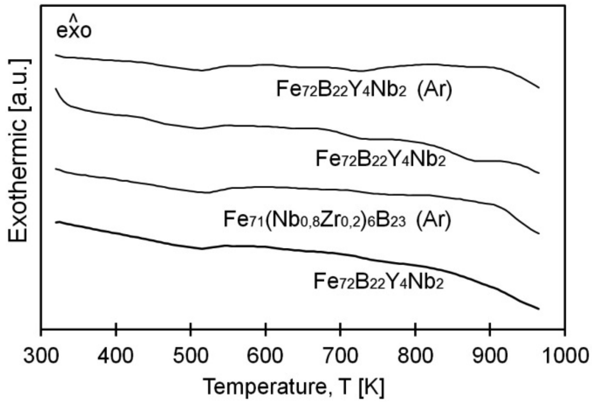

Fig. 6. DTA curves obtained from as-cast $Fe_{72}B_{22}Y_4Nb_2$ and $Fe_{71}(Nb_{0,8}Zr_{0,2})_6B_{23}$ alloys

Characteristic inflexion of the measurement line on the thermograms has not been identified. Calorimetric curves do not show structural changes that should occur in the test samples during the heat treatment. Despite the increase of temperature derivatograms do not change their position in the exothermic direction.

3.3. Microscopic observation

The optical microstructure of the $Fe_{72}B_{22}Y_4Nb_2$ and $Fe_{71}(Nb_{0,8}Zr_{0,2})_6B_{23}$ alloy plates obtained by the pressure die casting method in an argon atmosphere and without a protective atmosphere are presented in Figs. 7-16. The photos of the obtained structures are shown in figures in x400 and x1000 magnification. The optical microscope tests confirmed the formation of the crystalline structure in all samples. The structure is rather fine-grained in these alloys. Figures 11-14 show the equiaxial and round grains frequently.

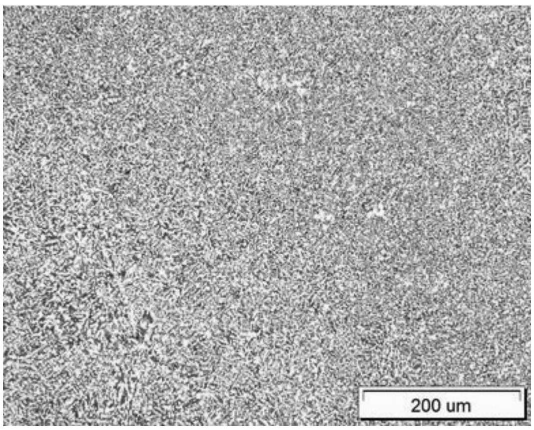

Fig. 7. Image of the microstructure of $Fe_{71}(Nb_{0,8}Zr_{0,2})_6B_{23}$ alloy obtained by die pressure casting method without protective atmosphere, the sample surface

The cross-sections of the samples are presented in Fig. 15. and Fig. 16. The structures of the cross-sections of the plates and the structures of the surfaces of the plates differ. The structures of the surfaces of the plates are similar in shape to the dendrites. As well as that the structures of cross-sections of the plates are characterized by a spherical grains and dendrites. Different heat removal rate to the mold is the cause of occurrence of dominant acicular structures on the surface of samples and dominant spherical (globular) structure in the middle of the samples. The difference was caused by a different crystallization process in different parts of the sample. The results of microscopic observations clearly indicate the lack of amorphous structure in the studied samples.

Such materials, characterized by an acicular structure, are hard yet brittle. Such a structure is undesirable in metal alloys. Presumably, small impurities might cause nucleation of crystalline phases, and consequently form a crystalline structure in all samples.

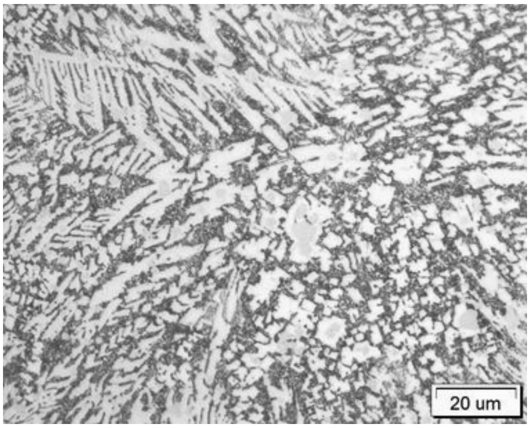

Fig. 8. Image of the microstructure of $Fe_{71}(Nb_{0,8}Zr_{0,2})_6B_{23}$ alloy obtained by die pressure casting method without protective atmosphere, the sample surface

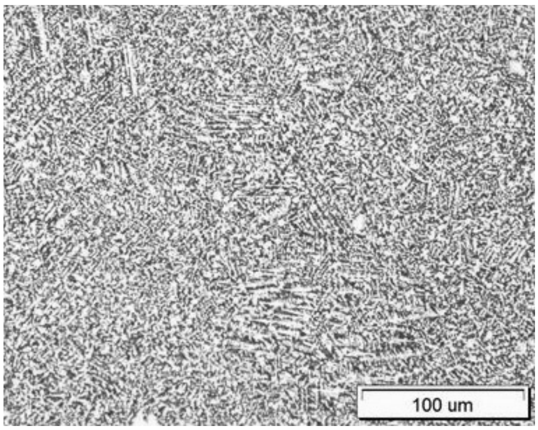

Fig. 9. Image of the microstructure of $Fe_{71}(Nb_{0,8}Zr_{0,2})_6B_{23}$ alloy obtained by die pressure casting method with protective atmosphere, the sample surface

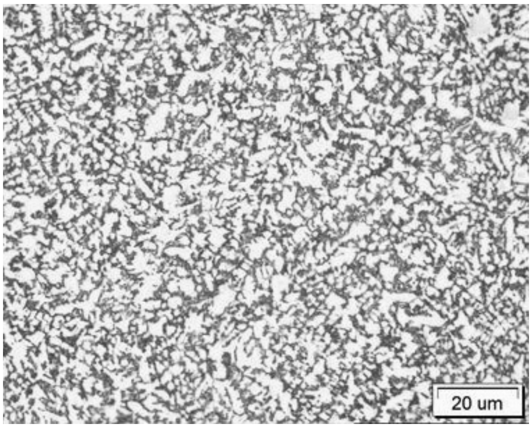

Fig. 10. Image of the microstructure of $Fe_{71}(Nb_{0,8}Zr_{0,2})_6B_{23}$ alloy obtained by die pressure casting method with protective atmosphere, the sample surface

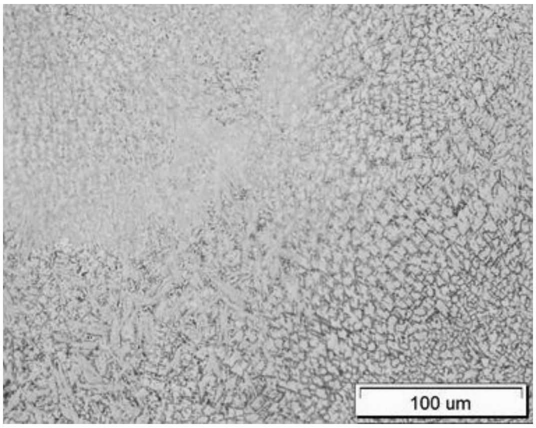

Fig. 11. Image of the microstructure of $Fe_{72}B_{22}Y_4Nb_2$ alloy obtained by die pressure casting method without protective atmosphere, the sample surface

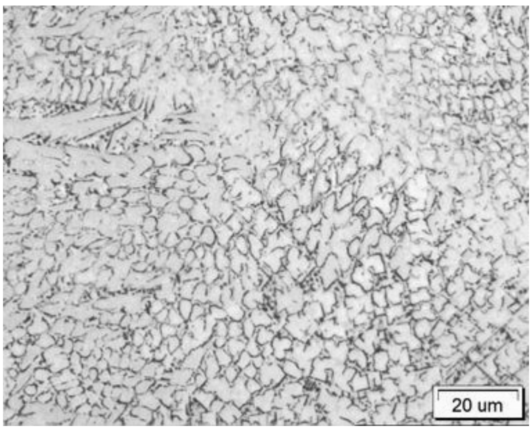

Fig. 12. Image of the microstructure of $Fe_{72}B_{22}Y_4Nb_2$ alloy obtained by die pressure casting method without protective atmosphere, the sample surface

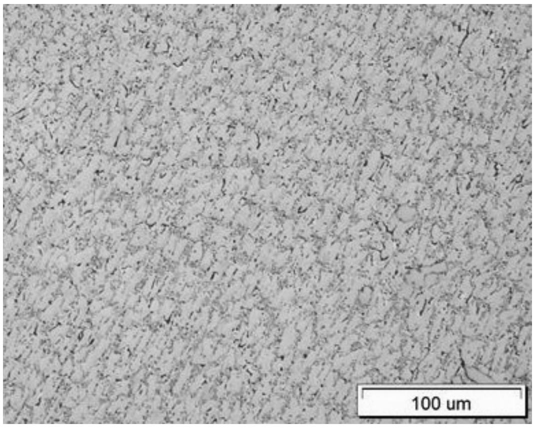

Fig. 13. Image of the microstructure of $Fe_{72}B_{22}Y_4Nb_2$ alloy obtained by die pressure casting method with protective atmosphere, the sample surface

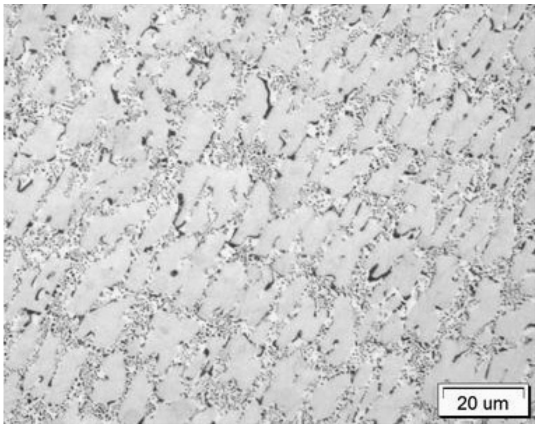

Fig. 14. Image of the microstructure of $Fe_{72}B_{22}Y_4Nb_2$ alloy obtained by die pressure casting method with protective atmosphere, the sample surface

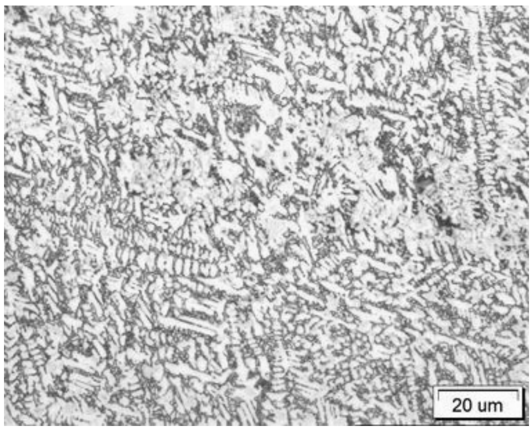

Fig. 15. Image of the microstructure of $Fe_{71}(Nb_{0,8}Zr_{0,2})_6B_{23}$ alloy obtained by die pressure casting method without protective atmosphere, cross section of sample

Fig. 16. Image of the microstructure of $Fe_{72}B_{22}Y_4Nb_2$ alloy obtained by die pressure casting method without protective atmosphere, cross section of sample

3.4. Hardness tests

Microhardness tests using the load equal to 1 kg was carried out. The Vickers hardness units were converted into the Rockwell hardness. Average hardness test results are shown in Fig. 17. The average hardness of the $Fe_{72}B_{22}Y_4Nb_2$ bulk metallic alloy was 1279 and 1268 HV whereas the average hardness of the $Fe_{71}(Nb_{0,8}Zr_{0,2})_6B_{23}$ alloy was 889 and 936 HV, adequately. The study showed a high hardness in all samples (> 65 HRC).

The alloys with yttrium addition have higher hardness in comparison to the alloys with zirconium addition. The application of a protective atmosphere does not have an impact on hardness. Table 4 shows hardness tests results obtained via other researchers [5,11-15].

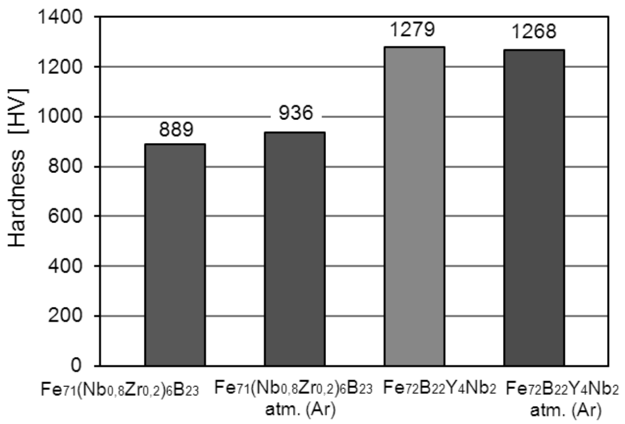

Fig. 17. Average hardness of Fe-Nb-B-Y and Fe-Nb-B-Zr system alloys

Fe-Nb-B-Zr system alloy samples exhibit lower hardness of about 100-150 HV in comparison to the literature results [5]. As well as that the hardness of Fe-Nb-B-Y alloy in comparison with the literature hardness results is higher by about 200 HV [11].

Tested alloys have higher hardness in comparison to Zr-based alloys [13,14]. The hardness of obtained samples is comparable to the required hardness of high-speed steel (at least 65 HRC) [15].

Table 4.
Hardness of chosen alloys - examples

Alloys	Hardness [HV]
$Fe_{74}Nb_6Y_3B_{17}$	1060
FeBZrNb	1030 ÷ 1290
FeCrNbB	1061 ÷ 1425
$Zr_{60}Al_{15}Ni_{10}Cu_{15}$	480
$Zr_{61}Al_{12.2}Ni_5Cu_{21.8}$	371
$Zr_{50}Cu_{40}Al_{10}$	580

4. Conclusions

The success of Fe-based metallic glasses production in form of plate is important for future progress of this group of materials. It is necessary mention that the higher dimension may extend the application temperature region of the bulk metallic glasses as engineering materials. In this research, the Fe-based alloy with Zr and Y addition in form of plate was produced by die pressure casting method.

It is more difficult to prepare a metallic glass of $Fe_{72}B_{22}Y_4Nb_2$ and $Fe_{71}(Nb_{0,8}Zr_{0,2})_6B_{23}$ alloys. Probably, the studied alloys are too far from the eutectic composition and it's very difficult to suppress the formation of dendrites phase. Perhaps, this is the reason why it does not exhibit an excellent GFA although it is a bulk glass-forming alloy. To obtain high GFA two aspect should be taken into consideration: stability of the liquid phase and resistance of the glassy phase to crystallization.

The lack of chemical heterogeneity control of obtained materials (ingots and plates) could be affected on the results of our research. There is necessity to the accurate uniform distribution of yttrium and zirconium in materials.

The investigation performed on the plates with thickness of 1 mm of Fe-Nb-B-Y and Fe-Nb-B-Zr system alloy allowed to formulate the following conclusions:

- the X-ray diffraction and differential thermal analysis investigations revealed that the studied alloys in as-cast state are crystalline. The use of protective atmosphere in the form of argon had no effect on the change of the structure which is also crystalline. The occurrence of following phases have been identified: α-Fe, Fe_2B, $Fe_{23}B6$, NbB,
- tests using an optical microscope revealed a dendritic and grainy structure. This structure is characteristic of crystalline alloys,
- the study of microhardness has shown high hardness in all samples (>65 HRC). The use of an argon atmosphere did not influence the results achieved. Alloys containing yttrium have a higher hardness than the alloys containing zirconium.

The studies exhibit that the obtained Fe-based bulk metallic alloys are not amorphous, but further and more precise production method is required.

Acknowledgements

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