Method of continuous casting of amorphous metallic materials based on iron

L. Jeziorski\textsuperscript{b}, M. G. Nabiałek\textsuperscript{a\textsuperscript{*}}, M. Szota\textsuperscript{b}, M. Dośpiał\textsuperscript{a}

\textsuperscript{a} Institute of Physics, Częstochowa University of Technology, 19 Armii Krajowej Av., 42 200 Częstochowa, Poland
\textsuperscript{b} Institute of Materials Science, Częstochowa University of Technology, 19 Armii Krajowej Av., 42 200 Częstochowa, Poland
\textsuperscript{*}Corresponding author. E-mail address: nmarcell@wp.pl

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Abstract

In this article, one of the production method of Fe based amorphous metallic materials, exhibiting excellent soft magnetic properties was described. The presented method consists in the unidirectional casting of the liquid alloy on to rotating, at high frequency, copper wheel. The produced samples had a ribbon shape with a thickness of about 45 microns. Apply this method allows to obtain materials exhibiting the lack of long-range ordering between the atoms and characterized by unique magnetic and mechanical properties. The structure of produced samples was investigated using Mössbauer spectroscopy and X-ray diffraction. The saturation magnetization ($\mu_0M_s$) and coercivity field ($H_c$) of ribbons in the as-cast state, have values corresponding to materials, with good soft ferromagnetic properties. On the basis of structural and magnetic investigations, it was found that the obtained samples were amorphous and had very good magnetic properties. That allows us to think, that the presented method makes it possible to fabricate the amorphous materials, characterized by the good electrotechnical parameters.

Keywords: Amorphous alloys, Melt-spinning method, Mössbauer spectroscopy, Saturation of the magnetization, Coercivity field

1. Introduction

The big problem of the industry are the world ending in coal deposits and environmental pollution. Therefore, industry is looking for modern, energy-saving and also environmentally safe functional materials. To this group of materials can be included the amorphous alloys. These alloys are materials with unique properties which are strongly dependent on their composition and fabrication process and it is the main reason for investigations performed by many of scientists, on their chemical composition and production methods. Very interesting amorphous alloys are ferromagnetic Fe-based materials, which exhibit very good soft magnetic properties, much better than the crystalline alloys with the same chemical compositions [1 ÷ 3]. Production of Fe-based materials, which are characterized by near-zero magnetocrystalline anisotropy as well as exhibiting low core losses, is very difficult to achieve. For production of such a material proper selection of manufacturing technique is required, what results from need to use enormous solidification speed of a liquid alloy. The most popular and widely used method of obtaining metallic glasses is a one-directional cooling of liquid alloy onto rotating, copper drum [4] and sometimes TIG method [5]. Cooling speed obtained during the continuous casting of ribbon reaches up to 10$^6$ K/s. Achieving such a large cooling rate of liquid metal during production makes it possible to obtain samples showing, in the entire volume, lack of recurrent configurations of atoms and angle-type correlation between them. Amorphous materials based on Fe in the form of thin ribbons are characterized by good soft magnetic properties and may in future
be used as material for construction of energy efficient and environmentally safe magnetic cores.

2. Research material and applied research techniques

Ingots were prepared using the high purity elements Fe – 99.98%, Nb - 99.999%, Y - 99.99%. Boron was added as an alloy with well known composition Fe₅₄,B₄₋₅₋₄. Samples were prepared using the melt-spinning method consisting of ultra-fast, one-directional cooling of a liquid alloy onto rotating copper cylinder. The obtained samples of Fe₇₃Y₃B₁₉Nb₅ alloy had the shape of ribbons with a thickness of 45 microns. The structure of manufactured samples was examined using transition Mössbauer spectroscopy and X-ray diffraction. The Mössbauer effect studies were carried out using "POLON" classical Mössbauer spectroscopy equipped with a ⁵⁷Co radiation source with an intensity of about 50 mCi and the half-life time of 270 days. X-ray diffraction patterns were obtained using "Dron 2" X-ray diffractometer equipped with a cobalt anode (λ₃₀ = 1,7902 Å). The samples were irradiated in the range of 2Θ from 30° to 120°, the angle increment equal to 0.05° and counting time of 3s. The magnetic properties of Fe₇₃Y₃B₁₉Nb₅ alloy were estimated on the basis of magnetic measurements carried out using a "LeakSchore" vibrating sample magnetometer, working in a magnetic field up to 2T and device for coercivity measurements basing on transformers method. The core losses were determined for frequencies from 50 Hz to 1000 Hz. All measurements were performed for samples in the as cast state and in the form of ribbons.

3. Fabrication method

One of the most popular method of producing amorphous, metal-based ribbons, is the method of continuous casting of a liquid alloy onto rotating, copper wheel. Smooth and stable stream of melt is directly targeted to the outer surface of the fast rotating copper drum, whose linear speed is usually equal of about 30 m/s. Cooling of the liquid alloy in this method is done through contact with the metal substrate, which very well transfers the heat. The liquid metal is collected on the outer surface of a rotating copper wheel in the form of the so-called "lake", from which the glassy ribbon is being pulled out (Fig. 1). Solidification of liquid metal takes place at very high cooling speeds, in the range from 10⁴ K/s to 10⁶ K/s. Cooling speed of an alloy squeezed out from quartz tube is the most important parameter characterizing this production method. In turn, cooling time is closely associated with the time during which the metal resides on a rotating at high frequency copper wheel. The discard of metallic ribbon is a result of centrifugal force action. Figure 1 shows a diagram describing the operating principle of the device for production of rapidly quenched tapes.

![Fig. 1. Diagram of the device for continuous casting of amorphous, metallic ribbons](image)

Devitrification process of liquid can be described using the schematic CTP curve (Fig. 2).

![Fig. 2. Schematic TCP curve describing the devitrification process: Sₓ - the cooling speed of liquid alloy onto the copper drum, Sᵧ - the cooling speed of liquid alloy in air, Sᵧ - critical cooling speed at which metallic glasses are obtained, tₓ - cooling time of metallic liquid , T₉ - glass transition temperature, Tₓ – melting point](image)

In Fig. 2 the schematic TCP curve is presented describing the glass formation process as well as cooling curve at a rate of (Sₓ) significantly greater than the critical cooling rate, the location on the TCP curve depends on the cooling time. If the cooling time of ribbon placed on the outer surface of the rotating copper drum would be too small to reduce the temperature below the melt temperature (Tₓ), the tape would be further cooled in the air at a rate (Sᵧ). That rate of cooling speed does not lead to formation of an amorphous state, as is clearly shown in Fig. 2. It follows from this, that devitrification process of liquid alloy, squeezed out from quartz tube under the argon pressure, is possible when the cooling time and cooling speed of the alloy (when the tape leaves the surface of the copper wheel) allows you to maintain a final temperature not exceeding the glass transition temperature (T₉). Effect of squeezing out pressure of liquid alloy onto copper drum, the linear velocity and size of hole located at the bottom of
the quartz tube on the fabrication of Co_{70.3}Si_{15}B_{10}Fe_{4.7} metallic ribbons is presented on Fig. 3.

![Fig. 3. Change of the rotation speed of the copper wheel as a function of hypertension.](image)

Boundary lines separating darkened area in Figure 3 are marked on the basis of experimental data set by Lachowicz [6], the dotted lines on the basis of [7]. Areas designated by gray and black circles show the results of experimental studies performed at the Institute of Materials Science and Engineering Warsaw University of Technology. Black dots specify the place where the experiment ended with the produce of the sample in the form of amorphous ribbon, white circles designate the places for which the production parameters did not provide the possibility of obtaining spin glasses. The experiment was performed for non-variable linear speed of the copper drum (v = 32 m / s), the hot melt temperature (T_m = 1200 °) and the same diameter of hole in a quartz capillary tube (0.22 mm). Fig. 3 shows that the argon pressure of squeezed out from quartz tube, molten alloy does not matter much for the thickness of obtained tapes.

4. Exemplary results of amorphous ribbons based on Fe and its discussion

Fig.4 presents X-ray diffraction pattern corresponding to the Fe_{73}Y_{3}B_{19}Nb_{5} alloy in the form of ribbon with a thickness of about 45 microns. In the case of crystalline materials, on X-ray diffraction pattern a presence of narrow peaks caused by the reflection of X-rays from the crystallographic planes (in these alloys are present long-range interaction between the atoms) is observed. In contrary, in amorphous materials because of the prevailing chaos in arrangement of atoms in the volume of the sample and increased density compared to crystalline materials with the same chemical compositions only short range interactions between the atoms are observed. Obtained diffraction pattern from the ribbon surface is typical as for amorphous materials. The broad, diffused maximum and absence of reflection peaks from the crystalline lattice testifies about the lack of repeated, atomic configuration in the alloy volume.

![Fig. 4. X-ray diffraction pattern of Fe_{73}Y_{3}B_{19}Nb_{5} amorphous alloy in the form of ribbon](image)

Amorphous structure in the as-cast ribbons was further confirmed by studies performed using ^{57}Fe Mössbauer spectroscopy. In Fig 5 transmission Mössbauer spectra are presented. These spectra are typical for materials with a disordered amorphous structure and are made up of the broad, asymmetric overlapping lines. Due to a changing surroundings of ^{57}Fe atoms, extending the absorption lines in the mössbauer spectra is observed (Fig. 5).

![Fig. 5. Mössbauer spectra of the Fe_{73}Y_{3}B_{19}Nb_{5} amorphous alloy in the form of ribbon with a thickness of 45 μm](image)

Fe-based amorphous materials with adapted chemical composition are often ferromagnetic alloys, showing a good, the so-called soft magnetic properties. In the amorphous alloys due to the lack of crystalline structure magnetocrystalline anisotropy is not observed, which significantly affects the growth of the coercivity field (H_c). Static hysteresis loop obtained for an amorphous soft magnetic material should be characterized by a low coercivity (H_L) and high saturation magnetization (μ_0M) (Fig. 6). Very troublesome phenomenon is observed in the commonly used transformers, built on the basis of FeSi sheets transformer have large magnetostriction, which is manifested by the change in the dimensions of the transformer core with an intensity two times higher than the frequency of the electricity grid. In addition, core losses arising in this type of cores are the reason of the loss of much of the transmitted energy and increasing the temperature of that transformer. Metallic amorphous materials have near–zero
and magnetostriction and a much smaller core loss than the commercially used transformer based on FeSi sheets. This means that the amorphous alloys produced on the basis of iron and alloying additions have the potential to be a great alternative as the part of modern, environmentally safe and cost-efficient transformers.

Fig. 6. Static hysteresis loop measured for the Fe73Y3B19Nb5 as-cast alloy in the form of ribbon

The core losses measured in the frequency range from 50 Hz to 1000 Hz for exemplary, presented in this work, amorphous alloy in the ribbon form are illustrated in Fig. 7.

Fig. 7. Core losses for the Fe73Y3B19Nb5 amorphous alloy in the ribbon form with a thickness of 45 μm

4. Conclusions

Presented in this paper a method of production of amorphous ribbons consisting of one-directional, surface cooling of liquid alloy, squeezed out from quartz tube under the argon pressure, makes it possible to obtain functional materials with high capacity application. The presented results for the amorphous, ferromagnetic material in a ribbon shape, show that, these materials can be included in a group of alloys exhibiting the so-called soft magnetic properties. This group of rapidly cooled materials, on the surface of rotating with high speed copper cylinder, shows great potential for application as a highly functional materials. Ability to produce transformers with excellent properties (almost zero magnetostriction, small coercivity field, high saturation magnetization and zero core losses) on the basis of this type of alloys, can contribute to the improvement of the environment, reduce consumption of natural resources and reduce expenditure on electricity.

In summary, described in this work, method can be used to produce modern, highly functional materials in the form of amorphous ribbons, characterized by good, soft magnetic properties.

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