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Influence of Hf and Y Content on the Local Occurrence of Antiferromagnetic Interactions in Amorphous Fe-Based Alloys

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Fe-based, rapid-quenched alloys are characterized by good magnetic properties. For alloys with ferromagnetic properties, an important parameter is the saturation magnetization. Its value depends on many factors, including: chemical composition, content of ferromagnetic elements, and structure of the material. For alloys with an amorphous structure, there is a chaotic arrangement of atoms in the alloy volume. Locally, however, the atoms may be in an order close to the crystal structure. The distances between magnetic atoms in such areas determine the magnetization process of the alloy. These distances can be adjusted with various alloy additions. The study investigated the effect of the content of Y and Hf on the distance between magnetic atoms and the value of saturation magnetization. It was found that a higher content of Y (characterized by a longer atomic radius compared to Hf) reduces the distance between Fe atoms and causes a decrease in the value of saturation magnetization. This is related to the local presence of antiferromagnetic order.

topics: bulk amorphous alloys, antiferromagnetic, Mössbauer spectroscopy

1. Introduction

The rapid development of human civilization, especially in the field of electronics and electrical engineering, constantly forces manufacturers to use newer and newer materials. These materials must meet rigorous shape quality standards and constantly improved performance parameters. Nowadays, electricity is a strategic medium and limiting its consumption is crucial. Therefore, materials that can be used to produce energy-saving electronic and electrical devices are being sought. Such materials are obtained by mixing appropriate ingredients while maintaining the rigor of production and obtaining the expected structure, both material and magnetic structure. Commonly known crystalline materials do not promise further development, therefore work is underway to change their structure. The change in structure may be related to the fragmentation of crystal grains present in the material, a change in their easy magnetization direction, or the complete elimination of the crystal structure. In conventionally used transformer sheets, their properties are improved in the texturing process, where a Goss or cube structure is obtained [1]. Such sheets can also be classified in terms of the directionality of their magnetic properties into isotropic and anisotropic. The former show the same magnetic properties in all measurement directions. The latter, however, have one specified direction of easy magnetization. From the point of view of magnetic properties, anisotropic steels are much better, but they have much worse mechanical properties. Both transformer sheets are obtained by cold rolling. The main difference in these materials is the Si content, which is approximately 1% for isotropic steel and from 3.5 to 6.5% for anisotropic steel [2]. The most desirable in terms of operational profitability is a transformer sheet with increased Si content, despite the observed decrease in saturation induction. The reason for this is that the phenomenon of magnetostriction is reduced to almost zero, which significantly reduces the operating costs of transformers. The group of electrical alloys showing a negligible value of magnetostriction includes a significant number of amorphous alloys, which are commonly called amorphous [3, 4]. These types of materials do not contain crystal grains and do not require additional plastic processing. In the production process itself, amorphous alloys become an anisotropic material, which, if necessary, can only be subjected to an annealing treatment, also known as relaxation annealing. The production of this type of electrical materials takes place at very high cooling rates (10^2–10^6 K/s), which significantly reduces their thickness. A very important factor determining the magnetic properties is the chemical composition of the material produced. The basis for the production of amorphous materials for use in the production of transformers is the FeCoB matrix. Of course, at this point, it is necessary to mention nanocrystalline materials (not discussed in this study), which are created as a result of the thermal treatment of amorphous materials and have properties different from them. In the case of rapid quenching materials after production, it can be shown that the elements are not evenly distributed in their volume, which leads to disturbances in their magnetic properties. The main reason may be the occurrence of the Invar phenomenon [5] or the competition between ferro- and antiferromagnetic ordering.

The paper will present the results of Mössbauer tests performed for the bulk $Fe_{65}Nb_5Y_{5+x}Hf_{5-x}B_{20}$ ($x = 0, 1$) amorphous alloys after solidification.

2. Experimental procedure

The test samples were prepared in two stages. The first stage involved making ingots, and the second stage involved casting rapid quenched alloys. Amorphous plates were obtained from high-purity ingredients: Fe — 99.99 at.%, Co — 99.999 at.%, Y — 99.99 at.%, Hf — 99.9999 at.%. Boron was introduced in the form of an alloy with the chemical composition of $Fe_{45.6}B_{54.4}$. The alloy ingots were made using an arc furnace operating in an argon atmosphere. The alloy components were weighed, placed on a water-cooled copper plate and melted using an electric arc (350 A). The sample was melted three times on each side. Before each melting, pure titanium was first melted, which acted as an absorber of the oxygen remaining in the chamber. After final melting, the ingots were left to cool to minimize oxidation of the ingot surface. The thus obtained ingots weighing 20 g were cleaned mechanically and in an ultrasonic bath.

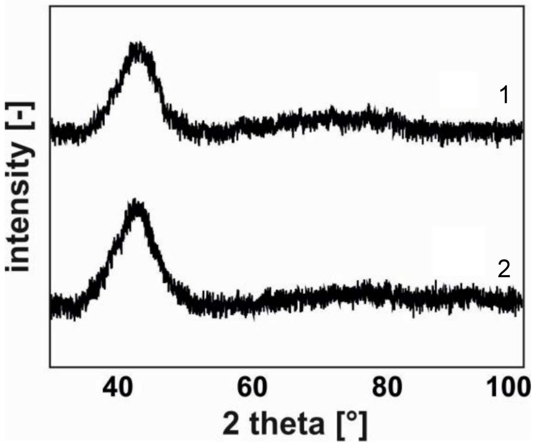

Fig. 1. X-ray diffraction images obtained for samples of the tested $Fe_{65}Nb_5Y_{5+x}Hf_{5-x}B_{20}$ alloys in the form of 0.5 mm plates: $x = 0$ (curve 1), $x = 1$ (curve 2).

After cleaning, they were crushed into smaller batch portions into an induction furnace, where bulk metallic glasses were produced. The plates were made in a chamber with a fixed argon atmosphere. Pieces of alloy melted using eddy currents were sprayed into a copper mold cooled by water. The liquid melt was solidified into a plate approximately 0.5 mm thick.

After solidification, the plates were subjected to X-ray diffraction (XRD) tests using a Bruker model ADVANCE 8 device. The device was equipped with a Cu X-ray tube, and the measurement was performed in the 2θ angle range from 30 to 100° with scanning density of 0.02° per measurement step and measurement time of 5 s. Mössbauer tests were performed on a POLON spectrometer. ^{57}Co was used as a Mössbauer source in a Rh matrix, with an activity of 50 mCi and a half-life of 270 days. The spectrometer was calibrated with 20 μm thick iron foil. Mössbauer spectra were recorded in transmission geometry. From the analysis of Mössbauer transmission spectra, the distributions of hyperfine field induction on ^{57}Fe nuclei were obtained. Primary magnetization curves were measured using a LakeShore vibration magnetometer operating at a magnetic field strength of up to 2 T. All measurements, i.e., XRD, vibrating-sample magnetometry (VSM) and Mössbauer measurement, were performed at room temperature.

3. Presentation of results

Figure 1 shows X-ray diffractograms obtained for samples of the tested alloys.

The X-ray diffractograms shown in Fig. 1 are similar and consist of a single broad maximum. This shape of the diffractograms indicates a chaotic arrangement of atoms in the sample volume for which

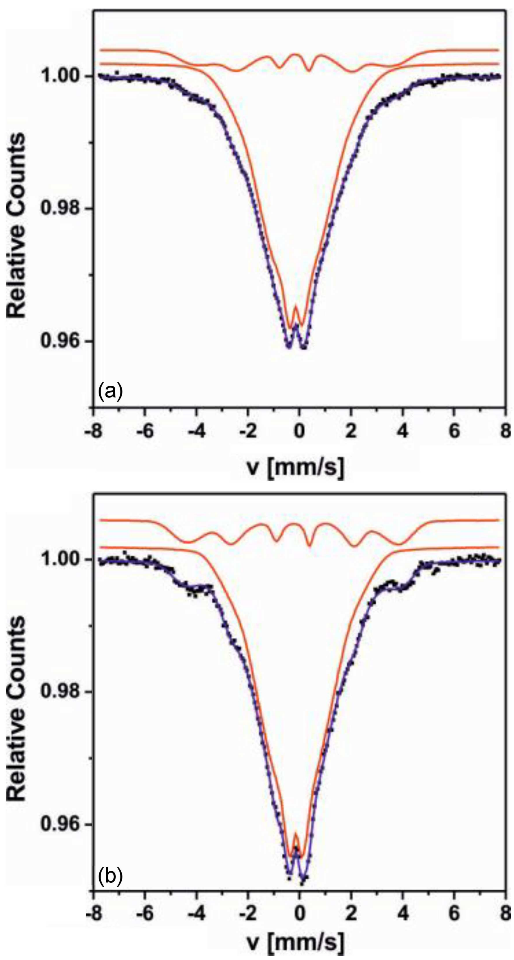

Fig. 2. Mössbauer transmission spectra obtained for the tested $Fe_{65}Nb_5Y_{5+x}Hf_{5-x}B_{20}$ alloys in the form of 0.5 mm plates: (a) $x = 0$, (b) $x = 1$.

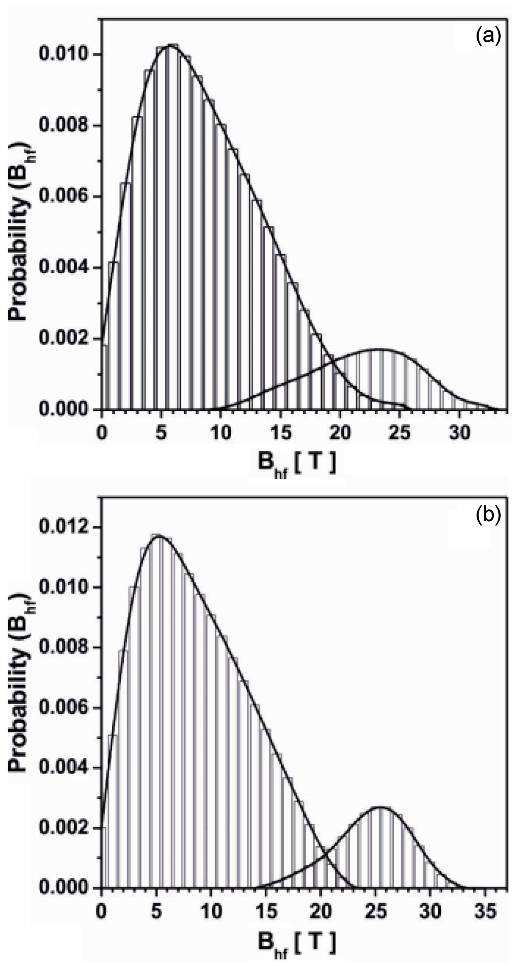

Fig. 3. Distributions of hyperfine field induction obtained based on the analysis of Mössbauer transmission spectra for the tested $Fe_{65}Nb_5Y_{5+x}Hf_{5-x}B_{20}$ alloys in the form of 0.5 mm plates: (a) $x = 0$, (b) $x = 1$.

there is no pattern, which in the case of crystalline materials gives an image in the form of a narrow peak related to the periodicity of the arrangement of atoms while maintaining the angular translations between them. The amorphousness of the samples was additionally confirmed by tests using Mössbauer spectroscopy. Figure 2 shows the Mössbauer transmission spectra measured for samples of the tested alloy after solidification.

The obtained Mössbauer spectra are typical for amorphous alloys. These spectra are asymmetric and consist of broad and overlapping lines. The so-called Zeeman sextets indicate that the tested material is an alloy with ferromagnetic properties. Based on the analysis of Mössbauer transmission spectra, the distributions of hyperfine field induction on ^{57}Fe nuclei were obtained (Fig. 3).

The distributions of hyperfine field induction on ^{57}Fe nuclei, obtained from the spectra (Fig. 2a and b), consist of two components, i.e., low field and high field. This shape of the presented distributions is related to the presence of different surroundings of iron atoms. As for the low-field component, it is associated with areas with high iron content. Increased concentrations of iron atoms force the distance between them to locally decrease, which may ultimately lead to the formation of antiferromagnetism. However, the high-field component is associated with iron atoms, which are also adjacent to Y and Hf atoms. For the sample with Y content = 5 (i.e, sample with $x = 0$), the average hyperfine field decreases, which probably leads to a local increase in the packing of Fe alloys. This state in the magnetic structure may manifest itself in the deterioration of saturation magnetization due to competition between ferro- and antiferromagnetic interactions. According to the Sleter–Bethe theory [6], reducing the distance between neighboring iron atoms results in a weakening of the ferromagnetic interactions and leads to a decrease in the hyperfine field of the low-field component in the hyperfine field distributions. This means that an increase in the share of the low-field component with a simultaneous decrease in its average hyperfine field may lead to the

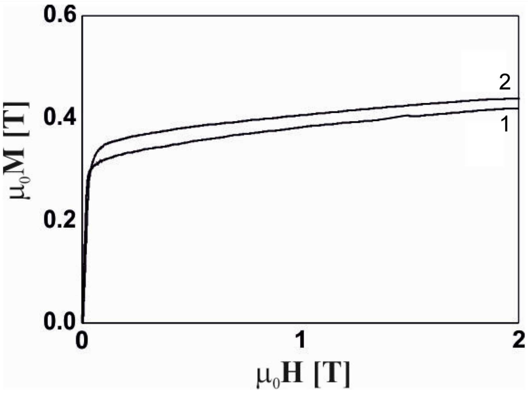

Fig. 4. Primary magnetization curves obtained for the tested $Fe_{65}Nb_5Y_{5+x}Hf_{5-x}B_{20}$ alloys in the form of 0.5 mm plates: $x = 0$ (curve 1), $x = 1$ (curve 2).

occurrence of areas with antiferromagnetic properties in the sample volume. Figure 4 shows the primary magnetization curves measured for the tested alloys.

For samples containing Y, a slight decrease in saturation is observed, which confirms the results of Mössbauer tests.

4. Conclusions

When designing amorphous alloys for use in electrical engineering, the phenomenon of rearrangement of ferromagnetic atoms in the sample volume should be taken into account. This rearrangement occurs already in the production process, which means that the most important element is developing the appropriate chemical composition of the alloy. As one can see, the introduction of elements with a large atomic radius has a negative impact on the homogeneity of the alloys after solidification. The observed increase in the low-field component with a simultaneous decrease in its average hyperfine field contributes to the competition between ferromagnetic and antiferromagnetic interactions.

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