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Influence of 1% Addition of Nb and W on the Relaxation Process in Classical Fe-Based Amorphous Alloys

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This paper features investigations into the influence of small additions of alloying elements on: structure, as well as the temporal and thermal stability of magnetic properties, and the disaccommodation effect, for the following amorphous alloys: $Fe_{61}Co_{10}Y_8Me_1B_{20}$ (where Me = Nb, W). The structure of the investigated samples has been confirmed by the Mössbauer spectroscopy. The obtained results indicate a strong correlation between the structure and the disaccommodation of the studied alloys. The Mössbauer studies reveal different configurations of atoms in the amorphous alloy samples, and the results indicate various potential barriers between orientations of atom pairs. For this reason, to describe the disaccommodation effect, the distribution of activation energy should be taken into account. The distribution of activation energy has been related to the distribution of relaxation times.

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1. Introduction

For the last 100 years, research has been undertaken into improvements in the magnetic properties of alloys used by the electrotechnical industry. In the beginning, to obtain magnetic material with so-called good soft magnetic properties required sophisticated production processes involving: directional rolling, thermal treatment, and gas supersaturation [1]. The resulting FeSi transformer steels were characterized by good properties; however they also exhibited high magnetostriction. In the 1960s, Duwez and his co-workers devised the production method for a new group of materials called the amorphous materials [2, 3]. Samples obtained by this process produces ribbon-shaped samples of approximate thickness 40 μm by rapidly cooling the molten alloy on a copper wheel at a rate of 10^4–10^6 K/s [2, 3].

Due to their exceptionally good properties, classical amorphous alloys, in the form of ribbons, are still intensively investigated in many research centres around the world [4–8]. In the amorphous materials, both short- and medium-range interactions exist between the atoms, and throughout the volume of the alloy, regions with different values of free energy can be distinguished. Systems like this are temporally unstable, and supplying the right amount of energy, the so-called activation energy, leads to an increase in their homogeneity, and in consequence — their crystallization [9]. Relaxation processes occurring during the production cycle or thermal treatment of amorphous alloys cause changes in the atomic order within the sample volume. The structural internal stresses, triggered by the presence of structural defects, influence the properties of this type of material [10]. In the amorphous materials, the structural defects can exist in the form of so-called "free volumes" and quasidislocational dipoles [11].

The structural defects in the amorphous materials, related with atomic discontinuities in the vicinity of the free volumes, could be investigated by measurement of the initial susceptibility (as a function of temperature) and its disaccommodation (MSD) [10].

The results of investigations, conducted by Kronmuller [11], showed that the disaccommodation of the magnetic susceptibility phenomenon is closely connected with the rearrangement of the atomic pairs in the vicinity of the free volumes, called also point defects.

In this work, the results of investigations are presented, revealing the effect of the addition of 1 at.% Me (where Me = Nb or W) on the relaxation processes in the classical amorphous alloys.

2. Experimental procedure

The initial ingots of $Fe_{61}Co_{10}Y_8W_1B_{20}$ and $Fe_{61}Co_{10}Y_8Nb_1B_{20}$ alloys were made by arc-melting high-purity component chemicals (≈99.99% pure). Boron was added to the initial ingot as an alloy with known composition — $Fe_{45.4}B_{44.6}$. The final samples were made in the form of ribbons by the melt-spinning method, using a linear velocity of 30 m/s. Both processes were completed under an inert argon atmosphere.

After the production process, the obtained samples were subjected to microstructural investigations, using the Mössbauer spectroscopy. Transmission Mössbauer spectra were obtained using a "POLON" spectrometer with an ^{57}Fe source of intensity approximately 50 mCi and half-life time of 270 days.

The low-field magnetic susceptibility and its disaccommodation were measured using a completely automated

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setup, based on a transformer method. All investigations were conducted on samples in the as-quenched state.

The magnetic after-effect was observed as a disaccommodation, i.e. a decrease over time of the initial magnetic susceptibility, following the demagnetization of the samples. The experimental results are presented as isochronal curves

$$\Delta(1/\chi) = 1/\chi_{120} - 1/\chi_2 = f(T), \qquad (1)$$

where χ_2 and χ_{120} are the susceptibilities measured at 2 s and 120 s after demagnetization of the sample [12].

3. Results and discussion

Figure 1 shows the Mössbauer spectra taken for the as-quenched, ribbon-shaped, samples of the $Fe_{61}Co_{10}Y_8W_1B_{20}$ and $Fe_{61}Co_{10}Y_8Nb_1B_{20}$ alloys. The Zeeman sextets are asymmetrical and consist of wide, overlapping lines. This shape of the Zeeman sextets is typical for ferromagnetic amorphous materials. Initial analysis of the obtained spectra showed

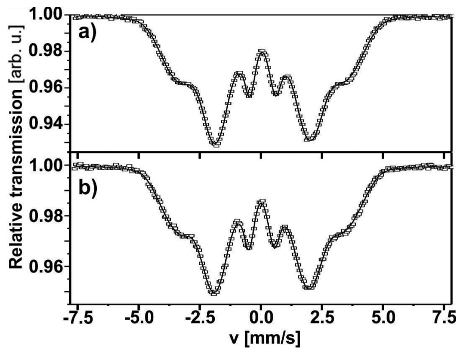

Fig. 1. Transmission Mössbauer spectra for the $Fe_{61}Co_{10}Y_8Me_1B_{20}$ alloys in the as-quenched state, where (a) Me = Nb, (b) Me = W.

that the inner lines in the presented spectrum are narrower than the outer lines which suggests a dominating distribution of the magnetic hyperfield B_{hf} in comparison to possible distributions of the remaining parameters [13]. The value of the relative intensity lines 2 and 5 (Table I) suggests spin arrangements along the axis of the ribbon. This type of spin arrangement is the result of the production process deriving the induced anisotropy.

TABLE I

The value of the mean hyperfine field using the ^{57}Fe nuclei (B_{ef}) and dispersion of the hyperfine field distributions of the amorphous phase (D_{am}), the relative intensity of lines 2 and 5 in the Zeeman sextet ($\langle A_{2,5}\rangle$).

Sample	B_{ef} [T]	D_{am} [T]	$\langle A_{2,5}\rangle$
Nb	19.39	4.995	3.31
W	19.19	5.004	3.15

From analysis of the Mössbauer spectra the hyperfine field distribution has been obtained (B_{hf}), as presented in Fig. 2. In the hyperfine field distribution B_{hf}, two clearly separated components could be distinguished: low- and high-field components. This indicates two different types of local configurations surrounding the Fe atoms [4, 5, 14].

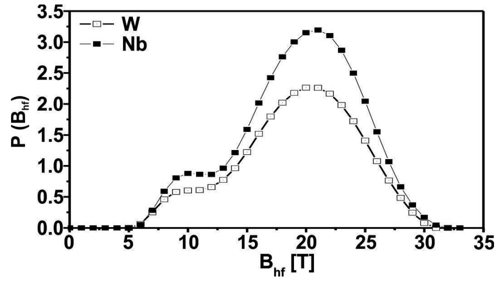

Fig. 2. Hyperfine field distributions for the $Fe_{61}Co_{10}Y_8W_1B_{20}$ $Fe_{61}Co_{10}Y_8Nb_1B_{20}$ alloys in the as-quenched state.

Due to the different character of the additional elements introduced to the base alloy, information of their influences on the shape of B_{hf} has been found.

The spin texture, specified by the relative values of the intensity lines 2 and 5, to the value of the intensity of the innermost lines in the Mössbauer spectra, is more homogeneous for the alloy with the addition of Nb. The parameters obtained from analysis of the Mössbauer spectra have been assembled together in Table I.

The higher value of B_{hf} has been found for the alloy with addition of Nb, which acts as a stabilizer for the structure [10]. The higher value of B_{hf} and lower value of D_{am} reflect the higher value of the atomic packing density of the material and its homogeneity with regard to the atomic configuration surrounding each central atom of ^{57}Fe.

The experimental isochronal curves of disaccommodation of the magnetic susceptibility have been presented in Fig. 3; their shape is typical for amorphous materials. The maximum of the disaccommodation curve for the sample of the $Fe_{61}Co_{10}Y_8Nb_1B_{20}$ alloy occurs at a higher temperature than for the alloy with addition of W.

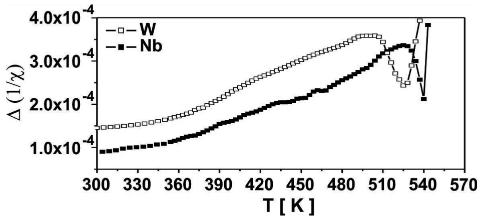

Fig. 3. The isochronal magnetic susceptibility disaccommodation curves $\Delta(1/\chi) = f(T)$ for the $Fe_{61}Co_{10}Y_8Nb_1B_{20}$ and $Fe_{61}Co_{10}Y_8W_1B_{20}$ ribbon samples.

Figure 4 shows the theoretical disaccommodation curves, with experimental points indicated. Numerical analysis has been performed according to the relationship [10]. The best fit has been found for the three elementary processes, each of which is de-

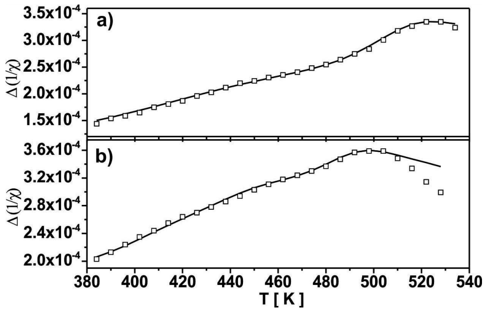

Fig. 4. Theoretical isochronal after-effect curve and experimental points obtained for the amorphous (a) $Fe_{61}Co_{10}Y_8Nb_1B_{20}$ and (b) $Fe_{61}Co_{10}Y_8W_1B_{20}$ ribbons.

TABLE II

Parameters obtained from fitting of isochronal disaccommodation curves for the amorphous $Fe_{61}Co_{10}Y_8Nb_1B_{20}$ and $Fe_{61}Co_{10}Y_8Nb_1B_{20}$ alloys in the as-quenched state.

Sample	Process	T_p [K]	I_p	Q_m [eV]	τ [10^{-15} s]
Nb	I	400	1.60 10^{-6}	1.27	2.45
	II	480	1.37 10^{-5}	1.52	2.48
	III	525	8.49 10^{-5}	1.65	3.94
W	I	440	5.90 10^{-5}	1.36	6.59
	II	460	5.31 10^{-5}	1.43	7.07
	III	505	6.34 10^{-5}	1.57	5.67

scribed by the Gaussian distribution of the relaxation times [10, 12].

The parameters obtained by the numerical analysis of the MSD curves have been collected in Table II, where I_p is the disaccommodation intensity at the peak temperature T_p, Q_m — average activation energies, and pre-exponential factor (τ) in the Arrhenius law.

The values obtained from the numerical analysis, according to Kronmüller theorem [11], showed that the relaxation times τ_0 are of the order of 10^{-15}, which suggests that the relaxation processes are related to the realignment of the axes of the atomic pairs in the vicinity of the free volumes. The lower value of the disaccommodation in the $Fe_{61}Co_{10}Y_8Nb_1B_{20}$ alloy reflects its higher value of time stability of the magnetic properties. This is connected with the lower number of point relaxators, which hinder atomic migration within the volume of the alloy.

4. Conclusions

The investigated ribbon-shaped samples of the alloys, $Fe_{61}Co_{10}Y_8W_1B_{20}$ and $Fe_{61}Co_{10}Y_8Nb_1B_{20}$, have been found to possess amorphous structure. This has been confirmed by investigations by means of the Mössbauer spectroscopy. On the basis of the obtained results it could be stated that the addition of Me in the form of Nb to the FeCoYMeB alloy, caused an increase in the average value of the hyperfine field induction and a shift in the maximum of the disaccommodation of the magnetic susceptibility towards a higher temperature. This behaviour of the B_{hf} and the MSD suggests a higher density of atomic packing in this material in comparison to the alloy with addition of W. Numerical analysis of the value of initial disaccommodation of the magnetic susceptibility showed that the pre-exponential factor τ in the Arrhenius law is of the order of 10^{-15}, which suggests that the structural relaxations in the investigated amorphous alloys are happening at the atomic level. In addition, the production process of the amorphous alloys solely promoted induced anisotropy in the sample, which was confirmed by the high value of the $\langle A_{2,5}\rangle$ coefficient, reflecting the spin texture.

In conclusion, within the amorphous alloys, even a slight change in alloy composition (1 at.%) could have a substantial influence on the resulting microstructure and magnetic properties. It is believed that these changes are connected with changes in the amorphous structure, which has a direct influence on the topological and chemical atomic order.

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