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

P. Pietrusiewicz1, K. Bloch1, M. Nabialek2 and S. Walters3

1Institute of Physics, Czestochowa University of Technology, Faculty of Production Engineering and Materials Technology, al. Armii Krajowej 19, 42-200 Czestochowa, Poland
2School of Computing, Engineering and Mathematics, University of Brighton, Cockcroft Building, Lewes Road, Moulsecoomb, Brighton BN2 4GJ, United Kingdom
3Department of Physics, University of Birmingham, Edgbaston, Birmingham B15 2TT, United Kingdom

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 hard magnetic properties required sophisticated production processes including: directional rolling, thermal treatment, and gas supersaturation [1]. The resulting FeSi transformer steels were characterized by magnetic properties; however, they also exhibited high magnetostriiction. 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 produce ribbon-shaped samples of approximate thickness 40 μm by rapidly cooling the molten alloy on a copper wheel at a rate of 107–109 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 quasidislocation dipoles [11].

The structural defects in the amorphous materials, related to 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 Kronmüller [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 Fe61Co10Y8W1B20 and Fe61Co10Y8Nb1B20 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 — Fe15.4B41.6. 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 a 57Fe 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...
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

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

where \( \chi_2 \) and \( \chi_{120} \) 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\(_8\)W\(_1\)B\(_{20}\) and Fe\(_{61}\)Co\(_{10}\)Y\(_8\)Nb\(_1\)B\(_{20}\) 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 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.

![Fig. 1. Transmission Mössbauer spectra for the Fe\(_{61}\)Co\(_{10}\)Y\(_8\)Me\(_1\)B\(_{20}\) alloys in the as-quenched state, where (a) Me = Nb, (b) Me = W.](image)

The value of the mean hyperfine field using the \(^{57}\)Fe nuclei \( B_{hf} \) 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 \( (\Delta A_{2,5}) \). 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\(_8\)Nb\(_1\)B\(_{20}\) alloy occurs at a higher temperature than for the alloy with addition of W.

![Fig. 2. Hyperfine field distributions for the Fe\(_{61}\)Co\(_{10}\)Y\(_8\)W\(_1\)B\(_{20}\) Fe\(_{61}\)Co\(_{10}\)Y\(_8\)Nb\(_1\)B\(_{20}\) alloys in the as-quenched state.](image)

![Fig. 3. The isochronal magnetic susceptibility disaccommodation curves \( \Delta(1/\chi) = f(T) \) for the Fe\(_{61}\)Co\(_{10}\)Y\(_8\)Nb\(_1\)B\(_{20}\) and Fe\(_{61}\)Co\(_{10}\)Y\(_8\)W\(_1\)B\(_{20}\) ribbon samples.](image)

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.

![Fig. 4 shows the theoretical disaccommodation curves, with experimental points indicated.](image)
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 ($\tau$) in the Arrhenius law.

The values obtained from the numerical analysis, according to Kronmüller theorem [11], showed that the relaxation times $\tau_0$ 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$_8$Nb$_1$B$_{20}$ 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$_8$W$_1$B$_{20}$ and Fe$_{61}$Co$_{10}$Y$_8$Nb$_1$B$_{20}$, 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 $\tau$ 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.

References