Magnetic Behaviour of Amorphous and Nanocrystalline $(\text{Fe}_{1-x}\text{Co}_x)_{86}\text{Zr}_6\text{B}_8$ ($x = 0$ and 0.5) Alloys

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The microstructure, and relationship between the magnetic properties and the measuring conditions of the amorphous and nanocrystalline Fe(Co)-Zr-B alloys were investigated. It was found that the volume fraction and their composition of the crystalline phase depend on the annealing conditions. The nanocrystalline $(\text{Fe}_{1-x}\text{Co}_x)_{86}\text{Zr}_6\text{B}_8$ ($x = 0$ and 0.5) alloys exhibit good magnetic properties with small amount of the crystalline phase.

Keywords: amorphous and nanocrystalline soft magnetic materials, Mössbauer spectroscopy, magnetic core loss.

1. Introduction

The nanocrystalline Fe-based alloys are an example of soft magnetic materials, which are important for technological applications and interesting from the basic research point of view. These alloys usually obtained by controlled crystallization of melt spun amorphous ribbons [1]. It has been found [2, 3] that the magnetic properties of the nanocrystalline alloys depend on preparation conditions of the amorphous ribbons. The procedure of the proper annealing allows one to obtain materials consisting nanocrystalline phase with ultrafine bcc-Fe grains of about 10-20 nm in size embedded in an amorphous matrix [4]. The good soft magnetic properties [5]-[9] these nanocrystalline materials are consequence of the reduction in the effective magnetocrystalline anisotropy on forming ultrafine grains [10]. Inoue *et al.* [11]-[13] have shown many interesting results of the investigations of the amorphous and nanocrystalline Fe-based alloys. The next investigations of the change in the microstructure and magnetic properties of the Fe(Co)-Zr-B alloys may give more details about their transition from the amorphous to the nanocrystalline state and explain role of Co addition to Fe-Zr-B alloys on their magnetic properties.

The aim of this paper is to study changes in the microstructure, and relationship between magnetic properties and the measuring conditions for the amorphous and nanocrystalline ribbons of the $(\text{Fe}_{1-x}\text{Co}_x)_{86}\text{Zr}_6\text{B}_8$ ($x = 0$ and 0.5) alloys.

2. Experimental procedure

The amorphous ribbons of the $(\text{Fe}_{1-x}\text{Co}_x)_{86}\text{Zr}_6\text{B}_8$ ($x = 0$ and 0.5) alloys were obtained by the rapid quenching

method. The ribbons of the $(\text{Fe}_{1-x}\text{Co}_x)_{86}\text{Zr}_6\text{B}_8$ ($x = 0$ and 0.5) alloys were 3 mm wide and about 0.028 mm thick. The nanocrystalline state was obtained by annealing of the amorphous ribbons in vacuum without a magnetic field. The microstructure of the samples was investigated at room temperature by Mössbauer spectroscopy (^{57}Co in a Rh source of 25mCi activity) and X-ray diffractometry with $\text{Co } K_\alpha$ radiation.

AC magnetic measurements for the $(\text{Fe}_{1-x}\text{Co}_x)_{86}\text{Zr}_6\text{B}_8$ ($x = 0$ and 0.5) alloys were performed using MMS-4001 (Ryowa Electronics Co., Ltd.) for 10 cm long ribbons.

3. Results and discussion

The amorphicity of the $(\text{Fe}_{1-x}\text{Co}_x)_{86}\text{Zr}_6\text{B}_8$ ($x = 0$ and 0.5) alloys were examined at room temperature by X-ray diffractometry. In the as-quenched state, all the investigated samples were fully amorphous and only a halo, which is characteristic for the amorphous materials was observed.

3.1 Samples of the $(\text{Fe}_{0.5}\text{Co}_{0.5})_{86}\text{Zr}_6\text{B}_8$ alloy

Fig. 1 shows Mössbauer spectra and hyperfine field distributions obtained for the nanocrystalline $(\text{Fe}_{0.5}\text{Co}_{0.5})_{86}\text{Zr}_6\text{B}_8$ alloy. The Mössbauer spectroscopy investigations show that the volume fraction of the investigated samples depends on annealing conditions.

Under the assumption that the average hyperfine field is proportional to the content of iron in the amorphous phase [14, 15], the volume fraction of the amorphous and crystalline phases were estimated from equations:

$$V_{am} = \frac{R \langle B_{am, asq} \rangle}{\langle B_{am} \rangle} \quad (1)$$

and

$$V_{cr} = 1 - V_{am} \quad (2)$$

where R is the relative area of the subspectra corresponding to the amorphous matrix, $\langle B_{am, asq} \rangle$ is the average hyperfine field obtained for the amorphous samples in the as-quenched state and $\langle B_{am} \rangle$ is the average hyperfine field obtained for the amorphous matrix.

The volume fraction of the crystalline phases in the investigated samples were estimated from difference

Fig. 1 Mössbauer spectra (a-c) and hyperfine field distributions (d-f) obtained for the nanocrystalline $(\text{Fe}_{0.5}\text{Co}_{0.5})_{86}\text{Zr}_6\text{B}_8$ alloy after annealing at 673 K for 30 min (a, d), 873 K for 10 min (b, e), and 873 K for 30 min (c, f).

between the total relative area corresponding to the crystalline phases and the relative area of the subspectra corresponding to the crystalline α -FeCo phase (confirmed by X-ray diffractometry). The phase composition of the crystalline phases was estimated from numerical analysis of the Mössbauer spectra (hyperfine field distribution) obtained for the investigated samples.

After annealing ribbon of the $(\text{Fe}_{0.5}\text{Co}_{0.5})_{86}\text{Zr}_6\text{B}_8$ alloy at 673 K for 30 min, the low (amorphous matrix) and the high (crystalline phases) field components in the hyperfine field distributions are observed. The average hyperfine field at ^{57}Fe nuclei (B_{eff}) and intensity of the second line in the Zeeman sextets ($A_{2,5}$) are equal 28.96 T and 2.73, respectively. After annealing of the sample of the $(\text{Fe}_{0.5}\text{Co}_{0.5})_{86}\text{Zr}_6\text{B}_8$ alloy at 873 K for 10 min, the small amount of the ferromagnetic amorphous matrix with high field component and crystalline phases were observed. In the crystalline component we can distinguish two phases: α -FeCo (72 % volume fraction) and $(\text{Fe}_{29}\text{Co}_{71})_{50}(\text{ZrB})_{50}$ (28% volume fraction). The parameters for this distribution are: $B_{\text{eff}} = 33.16$ T and $A_{2,5} = 2.67$. For the sample annealed at 873 K for 30 min only one component corresponding to the crystalline phases in the hyperfine field distribution was observed. In this alloys we can distinguish 78 % of the α -FeCo phase (with about 47 at% of Co atoms) and non-ferromagnetic $(\text{Fe}_{25}\text{Co}_{75})_{36}(\text{ZrB})_{64}$ phase. The intensity of the second lines in the Zeeman sextets is equal 2.45.

Fig. 2 Effective permeability versus frequency obtained for the $(\text{Fe}_{0.5}\text{Co}_{0.5})_{86}\text{Zr}_6\text{B}_8$ alloy in the as-quenched state under an applied ac magnetic field: 50 Am^{-1} (a), 100 Am^{-1} (b), 150 Am^{-1} (c), 200 Am^{-1} (d).

The structural changes occurring during annealing the amorphous alloys influences their magnetic properties. The Fe-based alloys in the as-quenched state exhibit rather poor soft magnetic properties. After proper annealing we can obtain materials, which characterize good soft magnetic properties.

Fig. 2 shows frequency dependence of the effective permeability for the $(\text{Fe}_{0.5}\text{Co}_{0.5})_{86}\text{Zr}_6\text{B}_8$ alloy. As can be seen from this figure the effective permeability keeps the constant values in the frequency range $10^2 - 3 \cdot 10^4$ Hz. Moreover, with increasing the magnetic field the effective permeability decreases.

When magnetic materials are magnetized by AC magnetic field $H = H_0 e^{i\omega t}$, the magnetic flux density B is generally delayed by the phase angle δ , and is thus expressed as $B = B_0 e^{i(\omega t - \delta)}$. The permeability is then [16]:

Fig. 3 Real and imaginary parts of the permeability versus the magnetic field for various frequencies: 240 Hz ($\circ$), 2 kHz ($\square$) and 10 kHz (Δ), for the $(\text{Fe}_{0.5}\text{Co}_{0.5})_{86}\text{Zr}_6\text{B}_8$ alloy in the as-quenched state.

Fig. 4 Relation between the core loss and frequency for the $(\text{Fe}_{0.5}\text{Co}_{0.5})_{86}\text{Zr}_6\text{B}_8$ alloy in the as-quenched state under an applied ac magnetic field: 50 Am^{-1} (a), 100 Am^{-1} (b), 150 Am^{-1} (c), 200 Am^{-1} (d).

$$\mu = \frac{B}{H} = \left(\frac{B_0}{H_0} \cos \delta - i \frac{B_0}{H_0} \sin \delta \right) = \mu' - i\mu'' \quad (3)$$

Fig. 5 Core loss versus frequency for the $(\text{Fe}_{0.5}\text{Co}_{0.5})_{86}\text{Zr}_6\text{B}_8$ alloy in the as-quenched state (a) and after annealing at 673 K for 30 min (b) and 873 K for 10 min (c), measured by applying a magnetic field of 200 Am^{-1} .

Fig. 6 Permeability curves for the $\text{Fe}_{86}\text{Zr}_6\text{B}_8$ alloy in the amorphous state: in as-quenched state (a) and annealed at 673 K for 30 min (b); and in nanocrystalline state: annealed at 873 K for 10 min (c) and at 873 K for 30 min (d), measured at a frequency of 2 kHz.

Fig. 3 shows real (μ') and imaginary (μ'') parts of the permeability versus the magnetic field for the $(\text{Fe}_{0.5}\text{Co}_{0.5})_{86}\text{Zr}_6\text{B}_8$ alloy in the as-quenched state. The real and imaginary parts of the permeability increase to a maximum at magnetic field range from 30 to 70 Am^{-1} , and then decrease. Moreover, the maximum values of the real and imaginary parts of the permeability shift to higher values of the magnetic field with increasing frequency.

Fig. 4 and Fig. 5 show the relation between the core loss and frequency for the $(\text{Fe}_{0.5}\text{Co}_{0.5})_{86}\text{Zr}_6\text{B}_8$ alloy under applying different magnetic fields and after different annealing conditions, respectively. For the investigated samples in the as-quenched state, with increasing the magnetic field from 50 Am^{-1} to 200 Am^{-1} , the core losses drastically increase. The core loss changes with annealing conditions. After annealing samples of the $(\text{Fe}_{0.5}\text{Co}_{0.5})_{86}\text{Zr}_6\text{B}_8$ alloy at 673 K for 30 min, the core loss above 1 kHz is smaller than for the samples in the as-quenched state and annealed at 873 K for 30 min. Moreover, for the higher frequencies, the eddy current dominates the losses.

Fig. 7 Permeability versus induction for the nanocrystalline $\text{Fe}_{86}\text{Zr}_6\text{B}_8$ alloy after annealing at 873 K for 30 min.

From obtained results and results presented in the next our paper [17], we can conclude that the partially crystallized $(\text{Fe}_{0.5}\text{Co}_{0.5})_{86}\text{Zr}_6\text{B}_8$ alloy shows tendency improve magnetic properties compared with the as-quenched state and the fully crystallized.

3.2 Samples of the $\text{Fe}_{86}\text{Zr}_6\text{B}_8$ alloy

Fig. 6 shows relation between the effective permeability and induction for the samples of the $\text{Fe}_{86}\text{Zr}_6\text{B}_8$ alloy. The effective permeability increases with increasing annealing temperature and time. The partially crystallized $\text{Fe}_{86}\text{Zr}_6\text{B}_8$ alloy (about 60% of the crystalline phase after annealing at 873 K for 30 min) exhibits the high effective permeability compared with the as-quenched state and after stress relieved (annealed at 673 K for 30 min). The maximum permeability for the samples of the $\text{Fe}_{86}\text{Zr}_6\text{B}_8$ alloy annealed at temperature 873 K for different time periods is for 0.58 T.

Relation between the effective permeability and induction for the ribbons of the $\text{Fe}_{86}\text{Zr}_6\text{B}_8$ alloy annealed at 873 K for 30 min are shown in Fig. 7. The effective permeability increases with induction and shift for the higher values of the induction. The maximum values of the effective permeability for all frequencies is about 11 000.

Fig. 8 shows the real and imaginary part of the permeability as a function of frequency for the $\text{Fe}_{86}\text{Zr}_6\text{B}_8$ alloy after stress relief and annealed at 873 K for 10 min. The cut off frequency for the ribbons of the $\text{Fe}_{86}\text{Zr}_6\text{B}_8$ alloy after annealed at 673 K for 30 min and 873 K for 10 min, where μ' has an inflection and μ'' has a peak in the frequency range from 100 Hz to 10 kHz were not observed.

Fig. 8 Real and imaginary parts of the permeability versus frequency for the samples of the $\text{Fe}_{86}\text{Zr}_6\text{B}_8$ alloy after annealing at 673 K for 30 min (squares), and at 873 K for 10 min (circles) under an applied magnetic field of 200 Am^{-1} .

4. Conclusions

The results obtained for the $(\text{Fe}_{1-x}\text{Co}_x)_{86}\text{Zr}_6\text{B}_8$ ($x = 0$ and 0.5) alloys with different amount of the crystalline phase are reported in this paper. The nanocrystalline materials of the $(\text{Fe}_{1-x}\text{Co}_x)_{86}\text{Zr}_6\text{B}_8$ ($x = 0$ and 0.5) alloys were obtained by annealing of the amorphous ribbons. It was found the after annealing the investigated samples the amount of the crystalline phase depends on the annealing temperature and composition. The microstructure of these samples influences their magnetic properties. The sample of the $(\text{Fe}_{0.5}\text{Co}_{0.5})_{86}\text{Zr}_6\text{B}_8$ alloy annealed at 673 K for 30 min consists α -FeCo phase embedded in the amorphous matrix and exhibits better magnetic properties compare with the as-quenched state. The nanocrystalline $\text{Fe}_{86}\text{Zr}_6\text{B}_8$ alloy annealed at 873 K for 30 min shows the high effective permeability and stability in the wide range of the frequency. All the samples partially crystallized are expected to be used widely in magnetic application fields.

5. References

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