ARCHIVES OF ACOUSTICS
18, 2, 311 – 321 (1993)

PIEZOMAGNETIC AND ULTRASONIC PROPERTIES OF THE Fe–Cu–Nb–Si–B METALLIC GLASS AFTER HEAT-TREATMENT

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An $Fe_{73.5}Cu_1Nb_3Si_{16.5}B_6$ alloy was cast as an initially amorphous ribbon. The samples about 50–51 mm long and 4 mm wide were cut out from the 10 mm wide and about 15 μm thick ribbon and annealed at the temperatures over 300°C (to 550°C) in the transverse magnetic field $H_\perp$ and also without magnetic field ($H=0$). Before nanocrystallization this alloy exhibits the saturation magnetostriction equal to $(20-23)\times 10^{-6}$. The annealing at the temperatures over 400°C in the transverse magnetic field increases the maximum value of the magnetomechanical coupling coefficient (k_m) from 0.10–0.15 (for as quenched state) to over 0.45. The magnetic anisotropy, introduced during the heat-treatment in the transverse magnetic field, is responsible for the predominant vector rotation mechanism which increases the magnetomechanical coupling and ΔE-effect.

1. Introduction

FINEMETs or Yoshizawa's alloys, i.e. iron-rich alloys with an ultrafine grain structure (with a diameter equal to 10–20 nm) are known as nanocrystalline materials [1–12]. These alloys after nanocrystallization are excellent soft magnetic materials, but before the crystallization they exhibit a saturation magnetostriction equal to $(20-23)\times 10^{-6}$ [1, 5–8] and they may be used as good piezomagnetic materials [9–11].

2. Experimental

An $Fe_{73.5}Cu_1Nb_3Si_{16.5}B_6$ alloy was cast as an initially amorphous ribbon [5–11]. The samples about 50–51 mm long and 4 mm wide were cut out from the 10 mm wide and about 15 μm thick ribbon and annealed at the temperatures over 300°C (to 550°C)

in the transverse magnetic field ($H_\perp$ = 650 kA/m) and also without magnetic field ($H=0$) [9–11]. In the first series the strips nos. 1–3 were annealed at the temperature of 350°C for 1/2 h in a transverse magnetic field. Next, the samples nos. 1 and 2 were annealed at a temperature of 400°C for 4 and 6 hours without magnetic field [9, 10]. In the second series these samples and the next one (i.e. nos. 1–4) were annealed at 500°C for 1/2 in $H_\perp$ and $H=0$, and at last at 550°C for 1/2 h without a magnetic field. In the third series next 11 samples (nos. 5–16) were annealed in transverse magnetic field for 1 h in the temperature range from 300 up to 500°C [11]. The characteristics of the magnetomechanical coupling coefficient (k) [13] and of the elasticity moduli at constant magnetic field (E_H) and at constant magnetic induction (E_B) versus magnetic bias field (H) were obtained using the resonance-antiresonance method [13–15]. The magnetomechanical coupling coefficient (k) was determined from the resonant (f_r) and antiresonant (f_a) frequencies of the half-wave length resonators, i.e. of the strip-shape samples [14, 15]

$$k^2 = \frac{\pi^2}{8}\frac{f_a^2 - f_r^2}{f_a^2}. \tag{1}$$

The moduli of elasticity at the constant magnetic field (E_H) and at the constant induction (E_B) were calculated also from these resonant and antiresonant frequencies, respectively, i.e.:

$$E_H = c_H^2 \rho \approx 4 l_r^2 f^2 \rho \tag{2}$$

and

$$E_B = c_B^2 \rho \approx 4 l_a^2 f^2 \rho \tag{3}$$

where l ($=\lambda/2$) is the length of the half-wave resonator ($l=50$ or 51 mm), ρ is density ($\rho=7.2$ Mg/m^3) and c_H and c_B are the ultrasound velocities at constant magnetic field H and at constant magnetic induction B respectively. The resonant frequencies were ranging from 30 to 52 kHz. The amplitude of the AC magnetic field was equal to 1–3 A/m. The characteristics of the impedance moduli are presented in Figures 1–3.

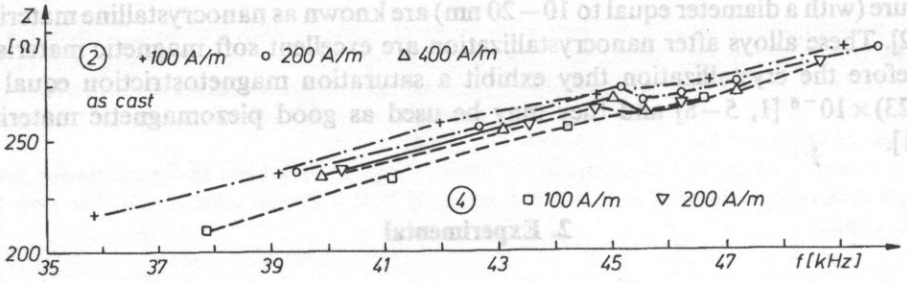

Fig. 1. Impedance (Z) vs. frequency (f) for samples no. 2 [10] and no. 4 in as-quenched state for magnetic bias field equal to 100, 200 and (400 A/m).

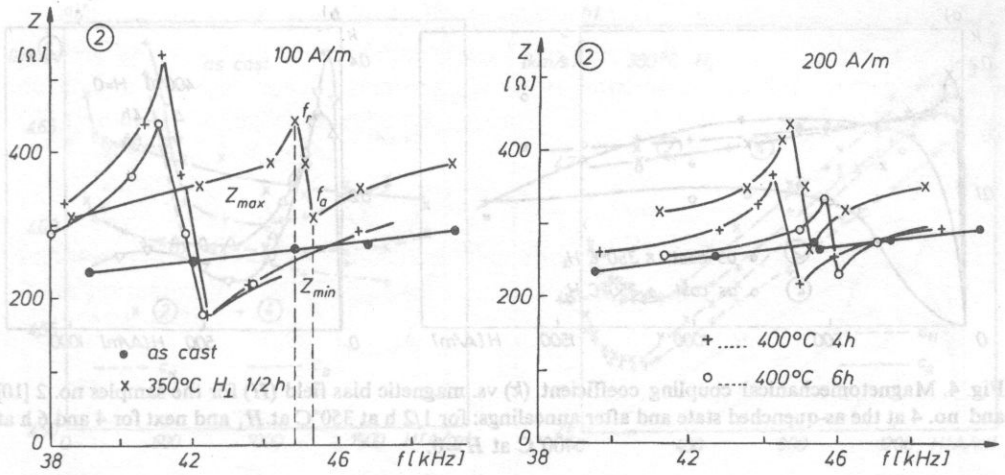

Fig. 2. Impedance (Z) vs. frequency (f) for sample no. 2 in as-quenched state and after annealing at transverse magnetic field ($H_\perp$ = 650 kA/m) for 1/2 h at 350°C and for 4 and 6 h without magnetic field at 400°C for magnetic bias field equal to 100 and 200 A/m [9].

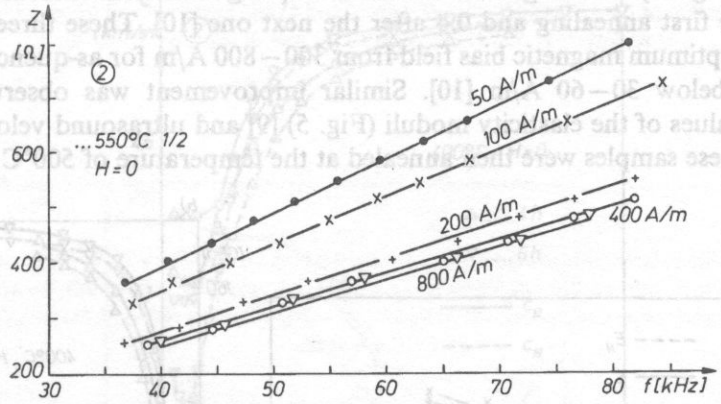

Fig. 3. Impedance (Z) vs. frequency (f) for sample no. 2 after last annealing for 1/2 h at 550°C and $H=0$.

Quasistatic hysteresis loops of magnetic induction (B) were measured and from these loops the coercive force (H_c) was determined [11]. Dynamical reversible magnetic permeability (μ) at the frequency of 10 kHz and the small amplitude of the magnetic field equal to 1A/m was measured versus quasistatic field [11].

3. Results and discussion

The annealing at the temperature of 350°C in the transverse magnetic field increases the maximum value of the magnetomechanical coupling coefficient (k) from

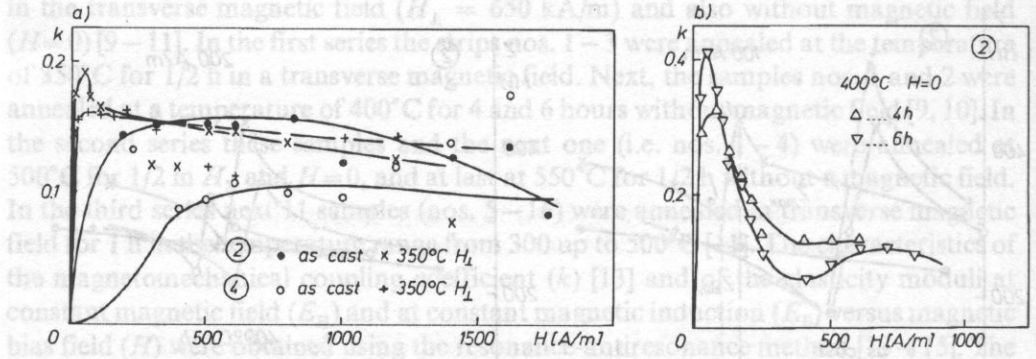

Fig. 4. Magnetomechanical coupling coefficient (k) vs. magnetic bias field (H) for the samples no. 2 [10] and no. 4 at the as-quenched state and after annealings: for 1/2 h at 350°C at $H_\perp$ and next for 4 and 6 h at 400°C at $H=0$.

0.10–0.15 to 0.15–0.19 (fig. 4a) [10]. The next annealings at the temperature of 400°C for 4 and 6 h without external magnetic field improved the piezomagnetic properties (Fig. 4b). The magnetomechanical coupling coefficient exceeds the value of 0.3 after the first annealing and 0.4 after the next one [10]. These three annealings shifted the optimum magnetic bias field from 300–800 A/m for as-quenched state to the values below 30–60 A/m [10]. Similar improvement was observed for the minimum values of the elasticity moduli (Fig. 5) [9] and ultrasound velocities (Figs. 6 and 7). These samples were then annealed at the temperature of 500°C for 1/2 h in

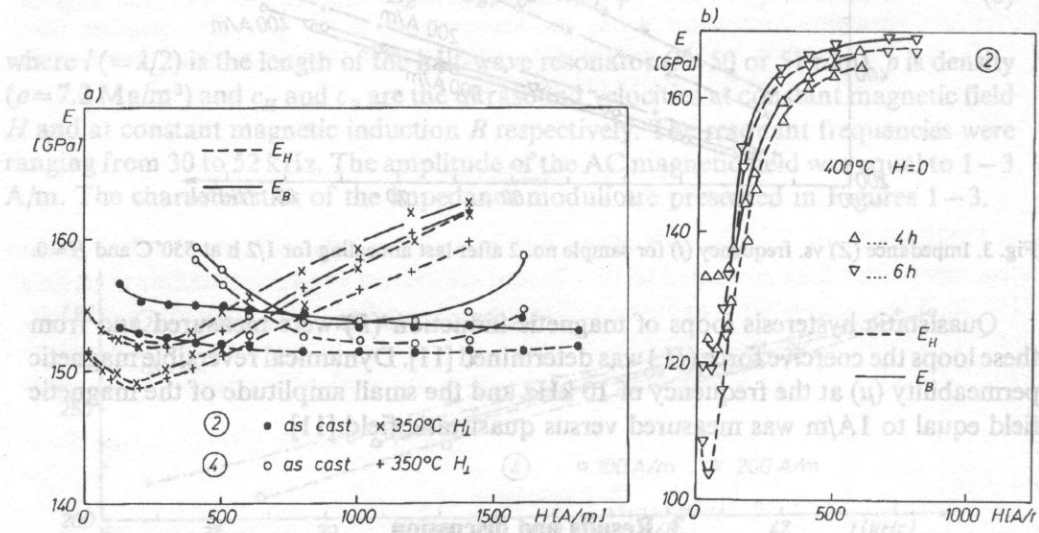

Fig. 5. Moduli of elasticity at constant magnetic field (E_H) and at constant induction (E_B) vs. magnetic bias field (H) for the as-quenched and annealed for 1/2 h at 350°C in $H_\perp$ samples (nos. 2 [9] and 4) and next annealed for 4 and 6 h at 400°C at $H=0$.

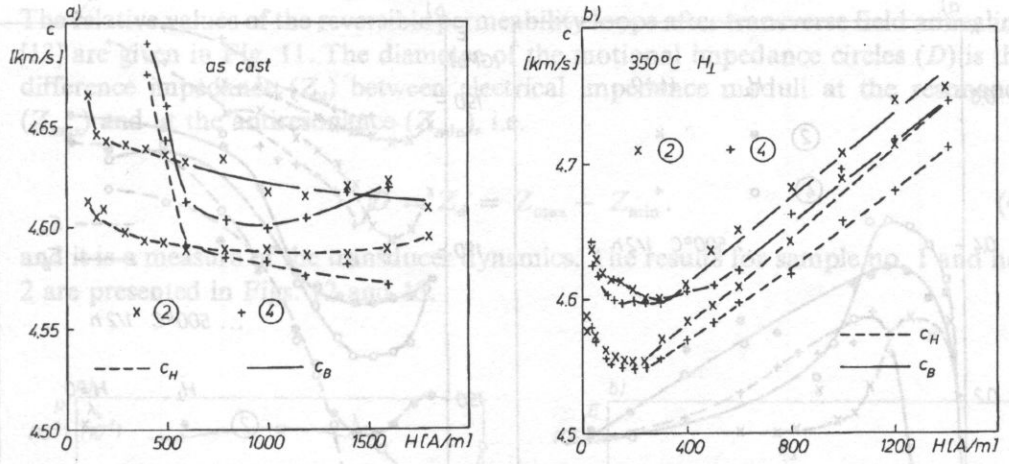

Fig. 6. Ultrasound velocities at constant magnetic field (c_H) and at constant induction (c_B) vs. magnetic bias field (H) for the as-quenched and annealed for 1/2 h at 350°C in $H_\perp$ samples nos. 2 and 4.

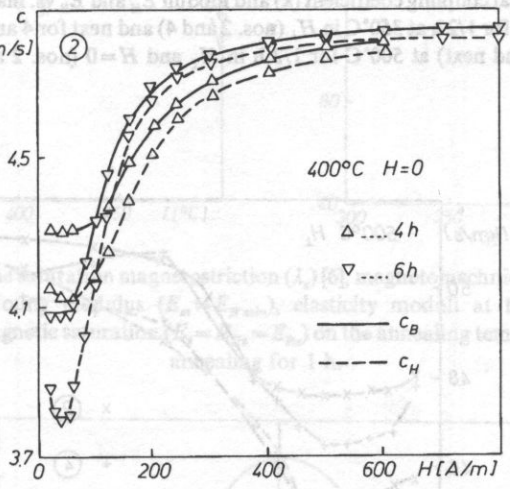

Fig. 7. Ultrasound velocities at constant magnetic field (c_H) and at constant induction (c_B) vs. magnetic bias field (H) for the as-quenched and annealed for 1/2 h at 350°C in $H_\perp$ 4 and next annealed for 4 and 6 h at 400°C at $H=0$ samples nos. 2 and 4.

the transverse magnetic field ($H_\perp$) and next without a magnetic field at 500 and 550°C for 1/2 h (Figs. 8, 9 and 3).

Summarized results from the third series of the investigations are presented in Fig. 10 [13]. The annealing temperatures starting from 300°C were increased to 320, 340, 360, 380, 400, 420, 440, 460, 480 and 500°C. An exception is the saturation magnetostriction (λ_s) for which the results were taken from the other papers [6–8].

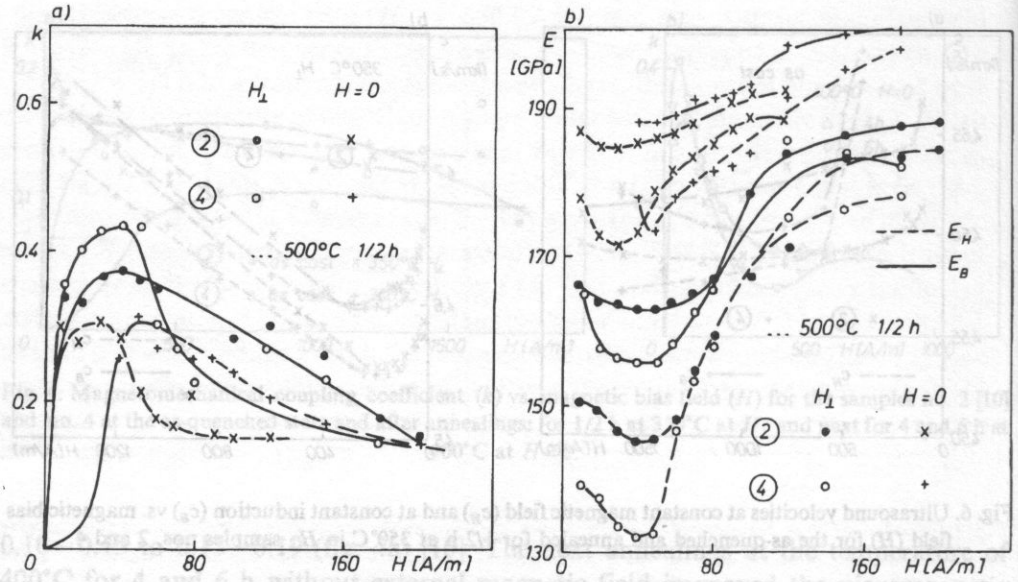

Fig. 8. Magnetomechanical coupling coefficient (k) and moduli E_H and E_B vs. magnetic bias field (H) for two samples after annealings: for 1/2 h at 350°C in $H_\perp$ (nos. 2 and 4) and next for 4 and 6 h at 400°C at $H=0$ (no. 2) and next) at 500°C for 1/2 h in $H_\perp$ and $H=0$ (nos. 2 and 4).

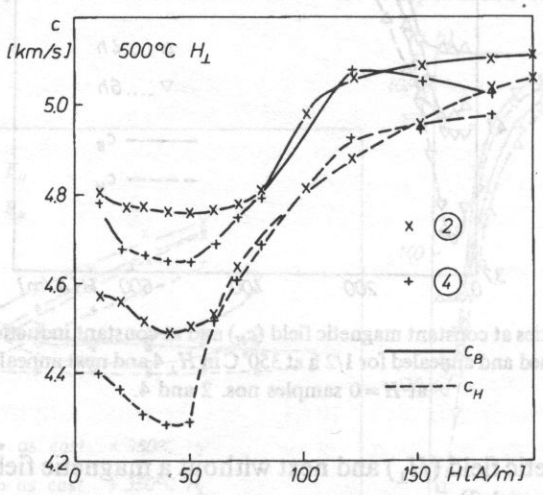

Fig. 9. Ultrasound velocities at constant magnetic field (c_H) and at constant induction (cE_B) vs. magnetic bias field (H) for two samples after annealings: for 1/2 h at 350°C in $H_\perp$ (nos. 2 and 4) and next for 4 and 6 h at 400°C at $H=0$ (no. 2) and next) at 500°C for 1/2 h in $H_\perp$ and $H=0$ (nos. 2 and 4).

The relative values of the reversible permeability loops after transverse field annealing [13] are given in Fig. 11. The diameter of the motional impedance circles (D) is the difference impedance (Z_d) between electrical impedance moduli at the resonance (Z_{max}) and at the antiresonance (Z_{min}), i.e.

$$D = Z_d = Z_{max} - Z_{min}. \tag{4}$$

and it is a measure of the transducer dynamics. The results for sample no. 1 and no. 2 are presented in Figs. 12 and 13.

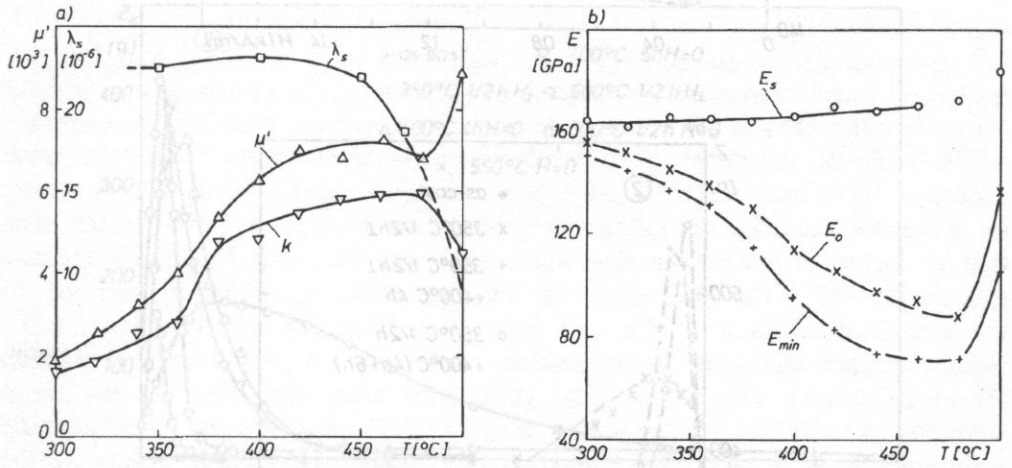

Fig. 10. Dependences of the saturation magnetostriction (λ_s) [6], magnetomechnical coupling coefficient (k), relative values of the Young modulus ($E_m = E_{H\,min}$), elasticity moduli at the demagnetization state ($E_0 = E_{H0} = E_{B0}$) and at magnetic saturation ($E_s = E_{Hs} = E_{Bs}$) on the annealing temperature of transverse field annealing for 1 h.

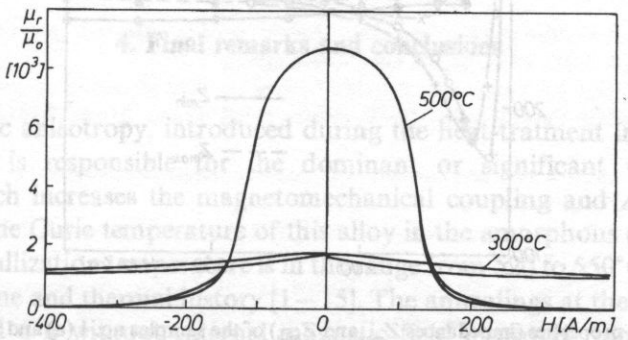

Fig. 11. Reversible ac permeability loops after transverse field annealings for 1 h at the temperature of 300 and 500°C.

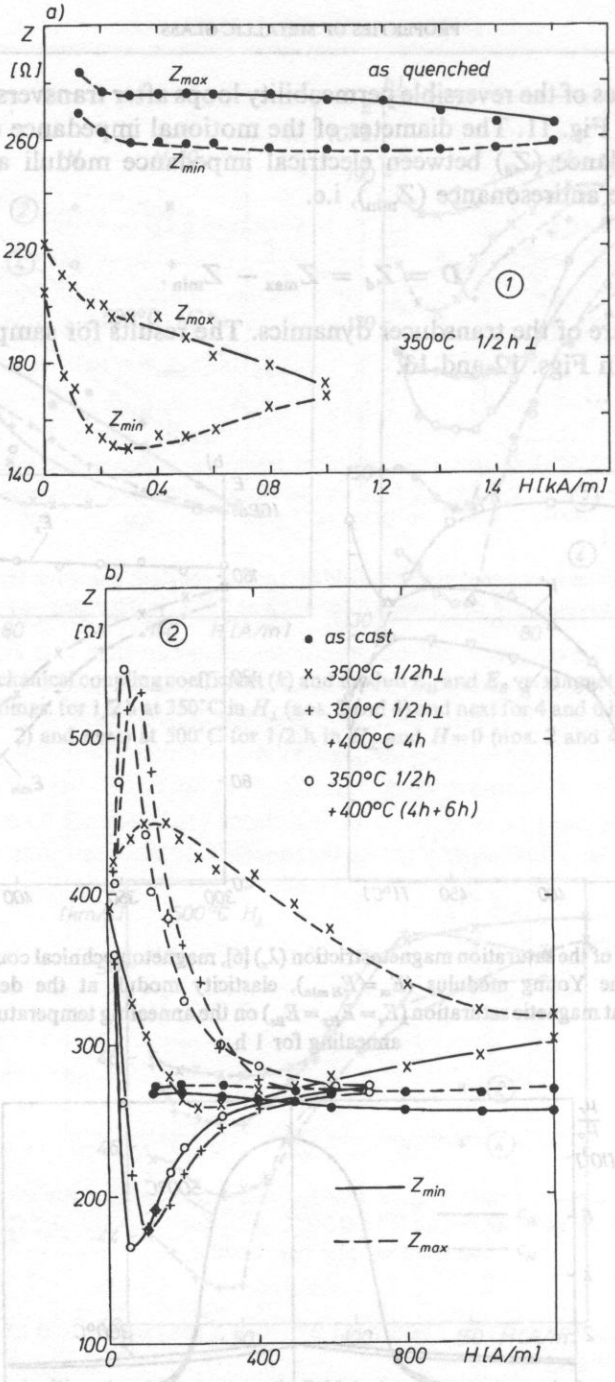

Fig. 12. Dynamical properties (impedances Z_{max} and Z_{min}) of the samples no. 1 (a) and no. 2 (b) vs. magnetic bias field (H) for as-casted state and after different heat treatment.

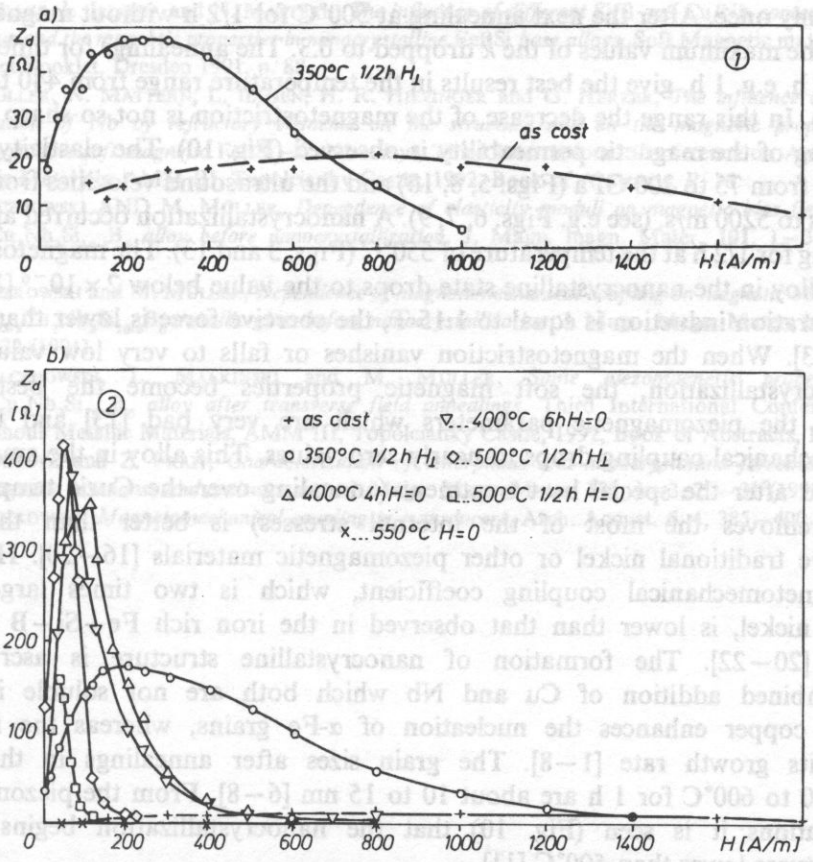

Fig. 13. Motional impedance $Z_d = Z_{max} - Z_{min}$ of the samples no. 1 (a) and no. 2 (b) vs. magnetic bias field (H) for as-casted state and after different heat treatment.

4. Final remarks and conclusions

The magnetic anisotropy, introduced during the heat-tratment in the transverse magnetic field, is responsible for the dominant or significant vector rotation mechanism which increases the magnetomechanical coupling and ΔE or Δc effect (Figs. 4–10). The Curie temperature of this alloy in the amorphous state is equal to 320°C and crystallization temperature is in the range from 520 to 550°C depending on the annealing time and thermal history [1–15]. The annealings at the temperature of 400°C for 4 and 6 h without external magnetic field improved the piezomagnetic properties (Figs. 4b, 5b and 7). After annealing at 500°C for 1/2 h in $H_\perp$ there was observed a decrease of the k coefficient for the samples with a longer thermal treatment history, and the increase of the k (up to 0.45) for the sample no. 4, annealed

before only once. After the next annealing at 500°C for 1/2 h without magnetic field ($H=0$) the maximum values of the k dropped to 0.3. The annealings for times longer than 1/2 h, e.g. 1 h, give the best results in the temperature range from 440 to 480°C (Fig. 10). In this range the decrease of the magnetostriction is not so sharp and the increasing of the magnetic permeability is observed (Fig. 10). The elasticity moduli changed from 75 to 200 GPa (Figs. 5, 8, 10) and the ultrasound velocities from about 3000 m/s to 5200 m/s, (see e.g. Figs. 6, 7, 9). A nanocrystallization occurred after next annealing for 1/2 h at the temperature of 550°C (Figs. 3 and 13). The magnetostriction in this alloy in the nanocrystalline state drops to the value below 2×10^{-6} [1, 5–8]. The saturation induction is equal to 1.15 T, the coercive force is lower than 1 A/m [1–8, 13]. When the magnetostriction vanishes or falls to very low values after thenanocrystallization, the soft magnetic properties become the best [1–8] but not the piezomagnetic parameters which are very bad [13] and the magnetomechanical coupling drops to nearly zero values. This alloy in the amorphous state and after the special heat treatment (annealing over the Curie temperature, which removes the most of the internal stresses) is better than the more expensive traditional nickel or other piezomagnetic materials [16–20]. However, its magnetomechanical coupling coefficient, which is two times larger than that of nickel, is lower than that observed in the iron rich Fe–Si–B metallic glasses [20–22]. The formation of nanocrystalline structure is ascribed to the combined addition of Cu and Nb which both are not soluble in α-Fe. Hereby copper enhances the nucleation of α-Fe grains, whereas the niobium lowers its growth rate [1–8]. The grain sizes after annealings in the range from 500 to 600°C for 1 h are about 10 to 15 nm [6–8]. From the piezomagnetic investigations it is seen (Fig. 10) that the nanocrystallization begins at the temperatures lower than 500°C [11].

References

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