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INVESTIGATION OF THE MAGNETIC PROPERTIES OF THE Ni77/Fe14.4/Cu4.5/Mo0.5/Cr2.2/Si0.2 ALLOY

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The positron annihilation and Mössbauer effect measurements in permalloy Ni77/Fe14.4/Cu4.5/Mo0.5/Cr2.2/Si0.2 for different cooling rates have been performed. The results have been discussed in view of the magnetic properties of the alloy. Based on the energy spectra of positron annihilation radiation and ^{57}Fe Mössbauer spectra, the positron polarization coefficient and the hyperfine field distributions have been calculated, respectively. We demonstrate that the results of both, positron annihilation and Mössbauer effect measurements, are in very close relation to the initial permeability $\mu_{0.4}$ (at $H = 0.4$ A/m) and resistivity measurements.

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1. Introduction

Recently many amorphous and nanocrystalline materials containing iron, as well as Ni–Fe systems, particularly those of the permalloy systems, have received considerable attention [1–5]. Low losses at high frequencies and a good thermal stability make permalloys promising candidates for soft magnetic applications. The heat treatment of the sample, in particular the annealing temperature and the cooling rate, is the basic problem from the point of view of excellent magnetic properties [6, 7]. The most representative result of the high temperature annealing is the change of the density of dislocations that influences coercivity. The best magnetic properties are usually attained by cooling with a specified rate in the temperature range of ordering. Nevertheless, the influence of the heat treatment on magnetic properties depends on alloy composition. Considering specified application in electric systems, the utility properties of the multi-component Ni-based alloys improve, whereas for binary alloys they get worse. In particular, for ternary NiFeMo alloys the distinct increase in the initial permeability $\mu_{0.4}$ (at $H = 0.4$ A/m), as well as the decrease in coercivity, is observed for annealing at 450°C [8]. Three and four-component alloys reveal also the anomaly of resistance. It concerns the drastic increase in the electric resistance due to ordering and it is associated with the characteristic decrease in the lattice constant [8]. Also the dependence of electrical resistivity, magnetization, coercive force, the distribution of ^{119}Sn hyperfine fields and the corresponding atomic order parameters on the heat treatment have been studied for ^{119}Sn-doped Ni_3Fe alloy [9, 10]. In the present paper we attempt to aid the results of the initial permeability ($\mu_{0.4}$) and resistivity (ρ) measurements in Ni77/Fe14.4/Cu4.5/Cr2.2/Mo0.5/Mn0.6/Si0.3 alloy (permalloy P77) by positron annihilation (PA) and Mössbauer effect (ME) methods.

2. Experimental details

2.1. Sample preparation, permeability and resistivity characteristics

The 10 mm wide and 0.2 mm thick ribbons of multi-component Ni-based alloy were prepared by melting of the nominal amount of alloy components in an arc furnace. The threefold roller method was applied; (i) hot-rolling of slugs, (ii) after annealing at 850°C (from 5 mm up to 0.8 mm thick) and finally (iii) cold-rolling (from 0.8 mm up to 0.2 mm thick) after annealing at 750°C. The final chemical composition of the alloy was determined as follows (at.%): Ni — 77.24%, Fe — 14.5%, Cu — 4.55%, Mn — 0.6%, Si — 0.22%, Mo — 0.68%, Cr — 2.31%, C — 0.005% and O_2 — 90 ppm. The ribbons were wounded onto toroidal cores and finally, the annealing of these specimens was carried out at $T_a = 1160$°C in an argon atmosphere. Four different cooling rates from the range 6–17°C/min were applied.

TABLE I
Parameters of electric resistance (ER) and magnetic permeability (MP) measurements in Ni77/Fe14.4/Cu4.5/Mo0.5/Cr2.2/Si0.2 alloy (permalloy P77).

Sample		ER	MP
Alloy code	Cooling rate [°C/min]	Resistivity ρ [Ω mm^2/m]	Initial permeability $\mu_{0.4}$ [G/Oe]
3863/939/19	6	0.628 (0.001)	60000
3863/941/24	11	0.629 (0.001)	58000
3863/931/11	13	0.623 (0.001)	53000
3863/932/13	17	0.635 (0.001)	47000

Considering specified application in electric systems, the ferromagnetic cores consisting of Ni77/Fe14.4/Cu4.5/Cr2.2/Mo0.5/Mn0.6/Si0.3 ribbons should have the initial permeability $\mu_{0.4} \geq 55000$ G/Oe. Only the slow cooled samples satisfied this condition. In general, the values of $\mu_{0.4}$ differ for different cooling rates. The resistivity has been estimated on the base of the well-known formula, $R = \rho l/s$, for 20 cm long pieces of ribbon, with accuracy of $\pm 10^{-3}$ Ω mm^2/m. Both, the results of $\mu_{0.4}$ and ρ measurements are shown in Table I.

2.2. Positron annihilation and Mössbauer effect measurements

The positron annihilation method has been applied to study of the ferromagnetic samples of Ni77/Fe14.4/Cu4.5/Cr2.2/Mo0.5/Mn0.6/Si0.3 alloy using the property of positron spin polarization in the external magnetic field. The standard gamma spectrometer with NaI(Tl) scintillators was employed. As a source of positrons ^{22}Na isotope of the activity of about 5 mCi was used. Both, the source and the sample were placed in an external magnetic field of $|B| = 600$ mT. The energy spectra of annihilation radiation for two opposite directions of the magnetic field were recorded in a multichannel analyser.

The ^{57}Fe Mössbauer spectra, for the same samples as in PA experiments, have been obtained at the room temperature using a conventional constant acceleration spectrometer. The transmission geometry and powdered samples have been applied. The monoenergetic 14.4 keV γ-ray was provided by the ^{57}Co source of the activity of about 50 mCi.

3. Results and discussion

The energy distributions of annihilation radiation, $I(E)$, have been measured for the two opposite (with respect to the source–sample direction) directions of the magnetic field. For $\boldsymbol{B}$ being parallel to the resultant spin of positrons emitted towards the sample, the spin of majority electrons in the sample is antiparallel to the positron spin ($S_e \uparrow\downarrow S_p$), whereas in the other case, i.e. $\boldsymbol{B}$ being antiparallel, the corresponding spins are parallel ($S_e \uparrow\uparrow S_p$). The measurements have shown that the area under the annihilation line in $I(E)$ distribution collected in a fixed time depends on the orientations of the magnetic field, i.e. the area for $S_e \uparrow\downarrow S_p$ (I_a) differs from the one for $S_e \uparrow\uparrow S_p$ (I_p), see Fig. 1. It is connected with the difference in cross-section for positron backscattering on electrons for parallel or antiparallel spin orientations and 3γ annihilation. 2γ annihilation occurs only if the spins of the annihilating positrons and electrons are aligned antiparallel. In the case of parallel spin alignment the 2γ annihilation is forbidden. 3γ annihilation is allowed but has a very low annihilation rate. To characterise the effect of the magnetic field on the positron annihilation radiation yield we introduced a parameter η (similarly as for angular distributions of annihilating quanta [11]), defined as

$$\eta = (I_a - I_p)/(I_a + I_p). \tag{1}$$

The corresponding values of η for different cooling rates are collected in Table II. As one can see, η shows the same character of change vs. cooling rate as ρ

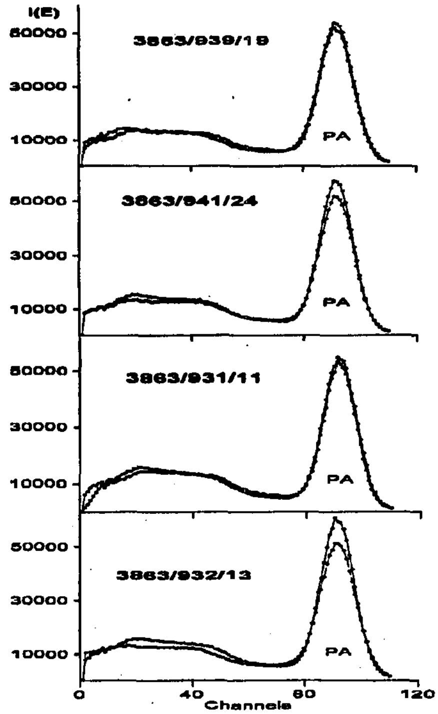

Fig. 1. Energy spectra of annihilation radiation for different cooling rates of permalloy P77 (PA — shows positron annihilation peaks for opposite orientation of external magnetic field; upper curves — parallel, lower curves — antiparallel to the resultant spin of emitted positrons, respectively; for more details — see text).

(see also Fig. 2). It is well known that the structural defects influence resistivity. As it was mentioned above, the multi-component NiFe alloys reveal anomaly of resistance consisting in the increase in resistance due to annealing in the temperature range of ordering. In consequence of ordering the distinct decrease in the lattice constant as well as creation of ordered Ni_3Fe compound are observed. Regarding the strong ferromagnetism of Ni_3Fe (magnetic moment in an ordered phase is greater than in the disordered one) such a situation is favourable to positron annihilation in defects (e.g. vacancies) in which the spin of electron corresponds to the direction of majority spin polarization. In general, from point of view of 2γ annihilation, in ordered state the number of defects in paramagnetic phase should relatively decrease but they become predominant in the ferromagnetic phase. As a consequence, the majority of positrons are trapped in defects whose electrons are spin-polarised antiparallel to the positron polarization. This means that polarised positrons can be used not only to study of the defects population, but also to detect the spin-polarization of electrons in such defects. The Mössbauer spectra of ^{57}Fe in the studied alloy have been analysed to yield hyperfine field distributions (HFD). In order to obtain reliable $P(B)$ distributions the fitting procedure

TABLE II

Parameters of the positron annihilation (PA) and Mössbauer effect (ME) measurements in Ni77/Fe14.4/Cu4.5/Mo0.5/Cr2.2/Si0.2 alloy (permalloy P77).

Sample		PA	ME
Alloy code	Cooling rate [°C/min]	Polarization coefficient η [%]	Average hyperfine field $\langle B \rangle$ [T]
3863/939/19	6	3.31 (0.15)	23.82 (0.01)
3863/941/24	11	3.62 (0.15)	23.89 (0.01)
3863/931/11	13	3.01 (0.15)	24.38 (0.01)
3863/932/13	17	4.61 (0.15)	24.36 (0.01)

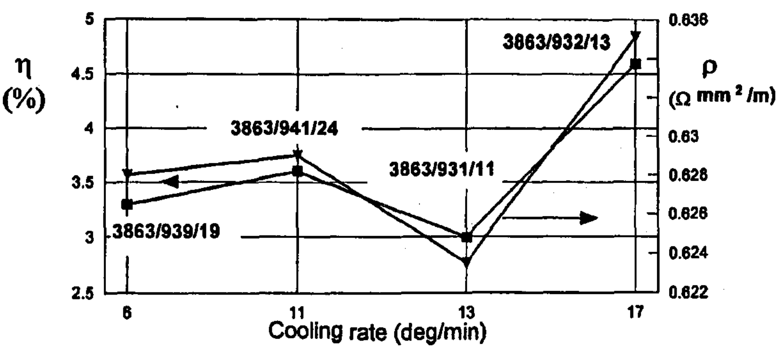

Fig. 2. Positron polarization coefficient (left scale) and resistivity (right scale) vs. cooling rate of permalloy P77.

has been employed to each spectrum in which the spectrum has been divided into low-field ($B_{lf} < 30$ T) and high-field ($B_{hf} > 30$ T) components. The method developed by Hesse and Rübartsch [12] in which a spectrum is fitted by a superposition of several sets of the six Lorentzians with different Zeeman splitting due to hyperfine fields with step of 1.0 T was used. The corresponding $P(B)$ distributions, as well as the values of the mean hyperfine field, $\langle B \rangle$, according to whole range of analysed $P(B)$ distributions are shown in Fig. 3 and in Table II, respectively.

As one can see from Fig. 3, the higher cooling rate produces an additional high field contribution to HFD and leads, in general, to more complicated magnetic structure of the alloy than in the case of slowly cooled samples. The growing up of HFD in the high field region (> 30 T) may result from disorder/order transition, in particular from ordered Ni_3Fe contribution to the six line pattern of the Mössbauer spectra of the alloy. In consequence, the mean hyperfine field rises (Fig. 4). It is worth noting that these results are in very close relation to $\mu_{0.4}$ measurements. As one can see from Table I, the highest values of $\mu_{0.4}$ were obtained for the cooling rates of 6 and 11°C/min. There are only two local maxima in $P(B)$ distribution associated to these samples (see also Table II) and it is clear

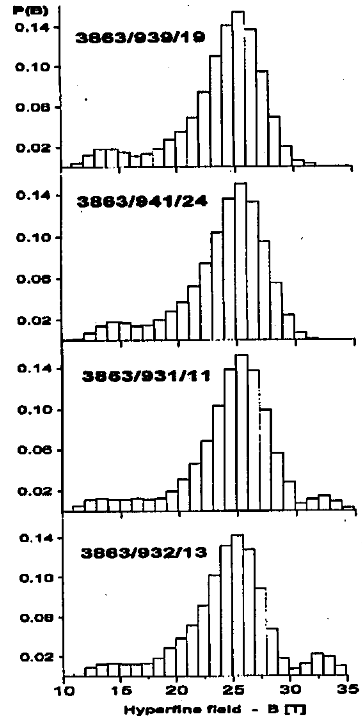

Fig. 3. The ^{57}Fe hyperfine field distributions for different cooling rates of permalloy P77.

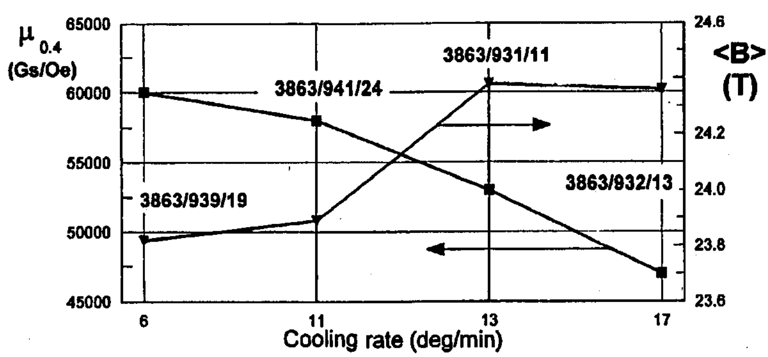

Fig. 4. Initial permeability (left scale) and the mean hyperfine field (right scale) vs. cooling rate of permalloy P77.

that the samples with the lower cooling rates remain better homogeneous from the point of view of the magnetic properties than the samples with the higher cooling rates (13 and 17°C/min). In magnetically inhomogeneous structure the motion of the domain walls is strongly depressed. It may be probably the main reason for variation of initial permeability from 60000 G/Oe (for the cooling rate of 6°C/min) to 47000 G/Oe (for the cooling rate of 17°C/min), see Fig. 4. However, the unexpected result has been obtained for the 3863/931/11 sample. As one can see from Table I and Table II, the resistivity as well as the positron polarization parameter are much lower than for other samples. It is clear that cold rolling should generate vacancies and dislocations. Therefore, the resistivity should increase, as well as the positron polarization parameter, because of the ability of positrons to be trapped in such defects. On the other hand, because of proportionality of positron polarization parameter to the average magnetic moment the change of η vs. cooling rate, similarly to the change of the mean hyperfine field ($\langle B\rangle \sim \mu_{\rm m}$), should be expected. Because of no special reason to consider the cooling rate of 13°C/min as an optimum, we suggest that the grains have been crumbled during preparation of the specimen. Also the result of the Mössbauer experiment seems to confirm the above conclusion. The nature of the Mössbauer spectra is determined to a considerable extent by the previous history of a sample. It was found by many authors [6, 7, 13, 14] that the mechanical, e.g. cold-working, and the irradiation treatments enhance the central peaks. It is also well known that the fcc structure in Fe–Ni alloys is stabilised in fine particles or by adding third elements such as carbon and manganese. In fine particles central peaks are enhanced. Even if the shape of the Mössbauer spectra is doubtful, the analysis of the HFD and the value of $\langle B\rangle$ can yield a valuable suggestion. In this case the central part of the Mössbauer spectrum is slightly broader and the $\langle B\rangle$ reaches the maximum.

In conclusion, at present we cannot explain the magnetic properties of the studied alloy in details, but it seems that the use of different techniques, including PA and ME, is a useful tool for investigation of the magnetic properties of permalloy materials.

Acknowledgments

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