MAGNETIC PROPERTIES OF THE Fe-Ni AND Fe-Ni-Mo SOFT MAGNETIC MATERIALS PREPARED BY THE MECHANICAL MILLING AND WARM CONSOLIDATION

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ABSTRACT

The aim of the present work was to study the influence of mechanical milling and subsequently the compacting on the structure and the soft magnetic properties of NiFe (81 wt. % of Ni) and NiFeMo (79 wt. % of Ni, 19 wt. % of Fe) alloys. We have investigated the influence of powder size on AC and DC magnetic properties of the bulk samples prepared by hot compaction.

Keywords: *soft magnetic material, mechanical milling, compaction, permalloy*

1. INTRODUCTION

Permalloy is the name which has been given to a series of nickel–iron alloys so heat-treated to have an initial permeability much larger than that of pure iron and are produced usually in the form of thin sheet (ring prepared by the wound ribbons or laminated thin sheets) [1].

Therefore it is logical to attempt to prepare such material direct in required form (more ‘‘bulk’’ form), for example in the form of a cylinder or a ring, which would be more convenient for some industrial applications.

Over the past several years the method of mechanical milling and mechanical alloying was widely spread in order to exploit it to produce a variety of equilibrium and non-equilibrium alloy phases and possesses further possibility for research work and application of permalloy. The advantage of this process technology is that the powder can be produced in large quantities and the processing parameters can be easily controlled [2]. One of the methods how to prepare material in bulk is to compact the powder. The aim of this work was to investigate the influence of powder size of Ni-based alloys of the bulk samples prepared by hot compaction on AC and DC magnetic properties.

The Ni-Fe based alloy (permalloy) system shows excellent soft magnetic properties and these alloys have been widely applied in the field of electronic devices and industry.

The aim of this work was to investigate the structure and magnetic properties of $Ni_{81}Fe_{19}$ (wt. %) and $Ni_{79}Fe_{16}Mo_5$ (wt. %) alloys and the contribution of the components of the core losses of bulk soft magnetic materials prepared by the compaction of alloy powder obtained by high-energy milling of the Ni-Fe ribbon.

2. SUBJECT

The soft magnetic properties of permalloy would change with Ni content: lower coercive field (about 80 at. % Ni), higher saturation magnetic induction (about 50 at. % Ni) and lower permeability but higher electrical resistance (about 35 at. % Ni) [3]. In [4] the effect of molybdenum alloying additions on the milling behaviour and subsequently on the magnetic properties was studied. It was found that molybdenum has a strong effect on the crystallization behaviour and NiFeMo alloys have very high relative permeability and low eddy current losses.

3. METHODS

We have prepared two types of powder samples (Fig. 1). The sample A was prepared by mechanical milling of microcrystalline ribbon NiFe (81 wt. % of Ni) obtained by melt-spinning. The ribbon is a good shape for milling. To prepare sample B, swarfs of NiFeMo (79 wt. % of Ni, 16 wt. % of Fe) were milled, which were prepared from the ingot by turning. We used swarfs, because it was not possible to prepare ribbons with this chemical composition. The milling of sample A and B was performed in protective argon atmosphere and in liquid nitrogen respectively in a high-energy planetary ball mill (RETSCH PM4000 with hardened steel vials and balls) with ball-to-powder-ratio of 6:1 and with a speed of 180 rpm.

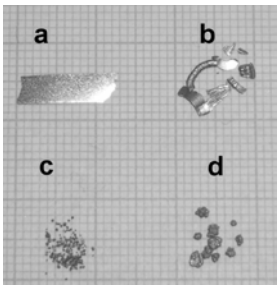

Fig. 1 Ribbon NiFe (a), powder prepared by milling of ribbon NiFe (c), swarfs NiFeMo (b), and powder prepared by milling of swarfs NiFeMo (d)

The bulk samples (Fig. 2) were prepared by uniaxial compaction of small pieces of broken ribbon (swarfs) with area of several mm^2 respectively of powders in the form of cylinders (diameter 10 mm, height 2.5 mm, weight approx. 2 g). The compaction was performed at a pressure of 800 MPa for 5 min at 600 °C in vacuum of 5×10^{-3} Pa (in order to prevent oxidation and to remove free gases

from powder before the compaction). The cylinders were annealed at temperatures between 500 °C and 1200 °C for 1 hour.

In order to prepare ring-shaped samples more suitable for AC and DC measurements, the cylinders of bulk samples were drilled using spark plasma erosion (the diameter is 5 mm, 10 primary turns, 20 secondary turns).

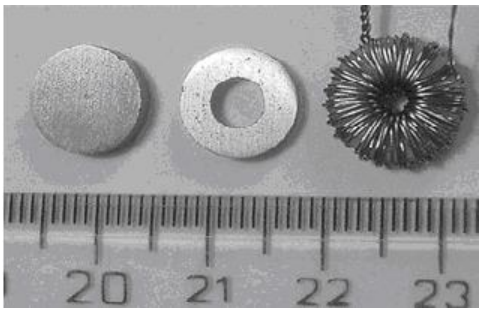

Fig. 2 Bulk samples $Ni_{81}Fe_{19}$ (wt.%) prepared for ac and dc measurements

4. RESULTS

4.1. Structure and morphology

The crystalline character of all samples (bulk, powder, NiFe, NiFeMo) was confirmed by X-ray diffraction with Fe filtered Co-Kα radiation (Philips PW 1050) with diffracted beam graphite monochromator [5] and differential scanning calorimeter (NETZSCH DSC 404), as we can see in Fig. 3 for sample NiFe and in Fig. 4 for sample NiFeMo. The reason for the different heat flow (Fig. 3) is the increasing of the energy, which was accumulated by the milling and then it released.

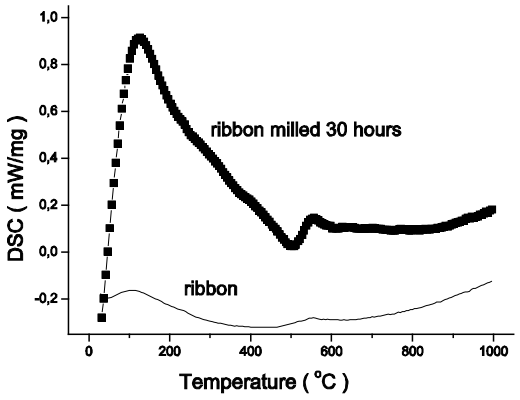

Fig. 3 DSC plots of samples $Ni_{81}Fe_{19}$

The $FeNi_3$ phase (T_C = 550 ºC) was created as major phase for Ni–Fe sample. The mechanical milling of Ni–Fe–Mo alloy causes the creation $FeNi_3$ phase and minor phase with higher content of Fe with T_C = 650 ºC [5].

The powder morphology and the morphology of the bulk samples were studied by scanning electron microscopy (Vega 5135 Tescan).

The morphology of the surface of the Ni–Fe ribbon in as-quenched state is depicted in Fig. 5a. Small pieces of the ribbon with area of several mm^2 were compacted, Fig. 5d. The ribbon pieces were brittle enough to be crushed during compaction process and the average linear size of the pieces was reduced to 5 μm. The 5 hours milled ribbon, consisting of powder elements with relatively sharp edges with size of about 30 μm (Fig. 5b), was precursor of the compacted bulk created by 3 μm elements, Fig. 5e. The powder milled for 30 hours had average size about 10 μm (Fig. 5c) and the surface of the bulk prepared from this powder is very smooth and no pores are visible, Fig. 5f [6].

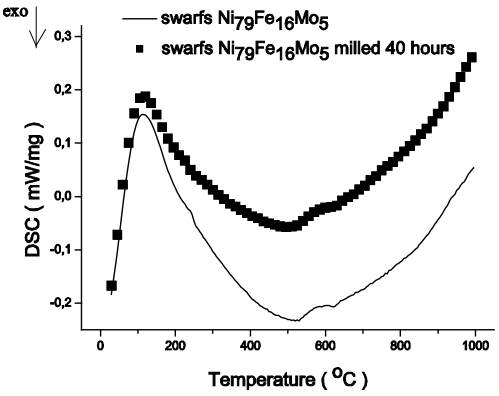

Fig. 4 DSC plots of samples $Ni_{79}Fe_{16}Mo_5$

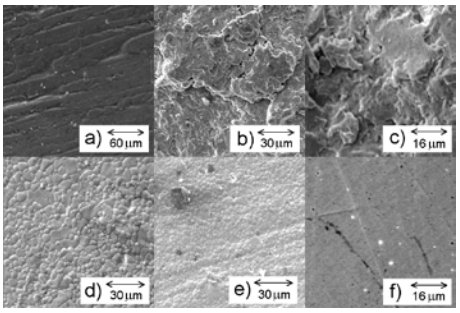

Fig. 5 The morphology a) of the Ni–Fe ribbon's surface, b) of the 5 h and c) of the 30 h milled powder, d) of the compact prepared of broken ribbon, e) of the compact prepared from 5 h milled powder and f) of the compact prepared from 30 h milled powder

4.2. Coercivity

The coercivity of the bulk samples was measured by a Forster Koerzimat at room temperature and is presented in Fig. 6 and Fig. 7 [6, 7].

The coercivity of the powder sample increases with milling time and we assume that displacement of the domain walls becomes less and less important magnetization process with milling time and the rotation of magnetization vector becomes more dominant. The magnetization process of the powder material is realized more or less separately for each powder element. The coercivity of the bulk material before heat treatment is lower than that for powder and that is why we can assume that the magnetic ''contact'' is restored after compaction. The annealing at higher temperatures causes relaxation of residual stresses introduced during milling and compaction and improves contact between powder particles, causing lowering of the coercivity. The lowest coercivity of 11 A/m (Fig. 6) was achieved for the sample NiFe prepared by compaction of broken ribbon, annealed

at 1200 ºC, and it is comparable with that for material prepared by convention way in the form of thin sheet 4 A/m [8].

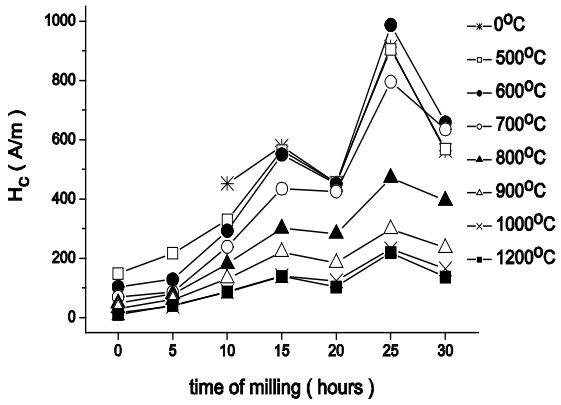

Fig. 6 Coercivity of bulk samples $Ni_{81}Fe_{19}$ milled for different time of milling (from 0 to 30 hours) and compacted at temperature to 1200 °C

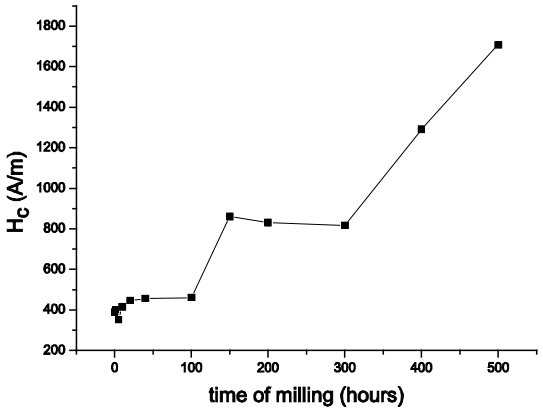

Fig. 7 Coercivity of bulk samples $Ni_{79}Fe_{16}Mo_5$ milled for different time of milling (from 0 to 550 hours)

4.3. Core losses

The losses were obtained from the ac hysteresis loops measured by a fluxmeter-based hysteresisgraph with sinusoidal flux density time and by the dc hysteresis loops. Peak permeability was determinated from B-H curves.

The core losses W (in J/m^3) in the magnetic cores can be divided into three components: hysteresis loss W_h, classical eddy current losses W_e and anomalous losses W_a [9]

$$W_t = W_h + W_e + W_a \tag{1}$$

Hysteresis losses W_h can be experimentally determined as the area of the dc hysteresis loop.

The eddy current losses W_e can be expressed as

$$W_e = \frac{\pi^2 B_{max}^2 d^2}{\beta}\sigma f = C_e f \tag{2}$$

where C_e is a constant in which the material parameters and the parameters of the measurement conditions except for the frequency are associated, d is the thickness of the sample, B_m is the maximum flux density, f is the frequency, σ is the conductivity, and β is a geometrical coefficient. For a rectangular cross-section perpendicular to the direction of the magnetic flux, the geometrical coefficient is

$$\beta = \frac{6}{1-0.633\left(\frac{w}{h}\right)\tanh\left(1.58\frac{h}{w}\right)} \tag{3}$$

where w is the width and h is the height of the rectangle [9].

The anomalous losses are known to be caused mainly by domain wall branching and bowing. These losses arise from the compensation of inhomogeneous internal counterfields (caused by eddy currents) by an applied magnetic field. The anomalous losses can be expressed as

$$W_a = \frac{8.8\sqrt{S}(B_{max})^{\frac{3}{2}}}{\rho}\sqrt{G\sigma V_0}(f)^{\frac{1}{2}} = C_a(f)^{\frac{1}{2}} \tag{4}$$

where C_a is a constant in which are associated the material parameters and the parameters of the measurement conditions except for the frequency, S is the cross section of the material perpendicular to the magnetic flux, G and V_0 are parameters that depend on the material and the magnetisation.

Fig. 8 and Fig. 9 show the peak permeability and the core losses, as a function of frequency for Ni–Fe–Mo and Ni–Fe samples for different values of the flux density B_{max}. The peak permeability is decreased, the core losses is increased.

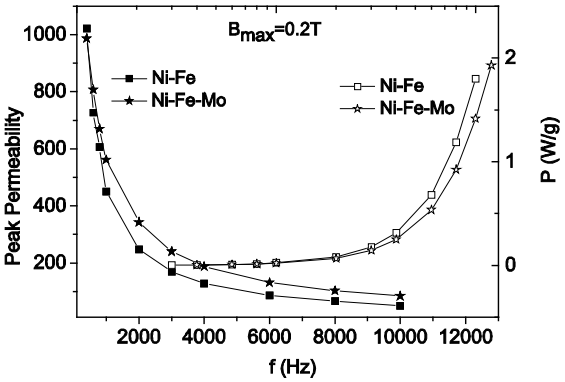

Fig. 8 Frequency dependence of the peak permeability and core losses for Ni-Fe sample and Ni–Fe–Mo sample for B_{max} = 0.2 T

Fig. 10 displays the dependence of the AC core losses and the AC coercivity in our bulk samples at a frequency of 20 kHz on the maximum induction, B_{max}. The core losses increase monotonically with the induction. As can be seen from this figure, the sample Ni–Fe–Mo is characterized by the lowest core losses, which is connected with the presence of Mo element, that slows down the ordering kinetics and lowers the degree of long

range order thereby increase the resistivity and simplifies the final heat treatment and hence improves the properties [10]. Table 1 compares the DC coercivity, AC core losses, and peak permeability in $Ni_{81}Fe_{19}$ bulk sample with $Ni_{79}Fe_{16}Mo_5$ bulk sample.

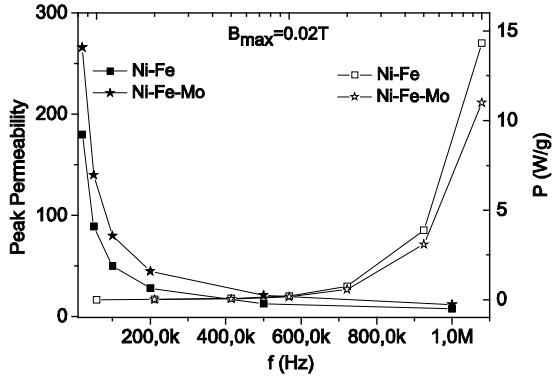

Fig. 9 Frequency dependence of the peak permeability and core losses for Ni-Fe sample and Ni–Fe–Mo sample for B_{max} = 0.02 T

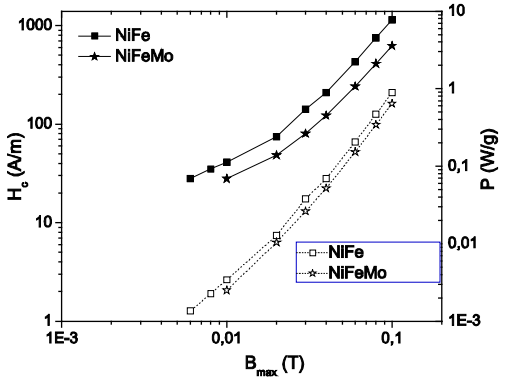

Fig. 10 Coercivity and core losses versus B_{max} at the frequency of 20 kHz for the Ni–Fe and Ni–Fe–Mo samples

Table 1 Magnetic properties of the prepared materials

	Ni-Fe	**Ni-Fe-Mo**
DC coercivity	11 A/m	11,2 A/m
Losses at f=10kHz, B_{max}=0,2T	1,8 W/g	1,4 W/g
Losses at f=1MHz, B_{max}=0,02T	14 W/g	11 W/g
Peak permeability at f=10kHz, B_{max}=0,2T	50	84

5. CONCLUSIONS

The soft magnetic properties of the $Ni_{81}Fe_{19}$ and $Ni_{79}Fe_{16}Mo_5$ alloys have been investigated. From the above study, we conclude that the magnetic properties of the permalloys and supermalloys (a metal alloy that is 79% nickel, 5% molybdenum and 16% iron) show strong dependence to their initial master powder and annealing conditions. The bulk sample $Ni_{79}Fe_{16}Mo_5$ is found to show better soft magnetic properties over the $Ni_{81}Fe_{19}$ bulk sample as a function of different process parameters. We have prepared bulk samples in the form of the small cylinders with coercivity down to 11 A/m. The discussed alloys show very good soft magnetic properties that can be tailored to the need of certain requirements by adjusting the chemical composition, the processing routes such as compaction, and heat treatment conditions. They have more degrees of freedom for tailoring their magnetic properties due to their flexibility in composition, shape and dimensions.

Therefore, to achieve very good soft magnetic properties of compacted-powder bulk materials, it is important to study the anomalous losses as a component of the total core losses.

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BIOGRAPHIES

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