MECHANICAL ALLOYING OF 80Ni-14.7Fe-4.4Mo-0.5Mn-0.3Si SOFT MAGNETIC PERMALLOY TYPE MATERIAL

H. Hadraba, M. Strečková, R. Husák, P. Roupcová

Abstract

80Ni-14.7Fe-4.4Mo-0.5Mn-0.3Si soft magnetic alloy of Permalloy type was prepared by mechanical alloying from atomic powders. The powders were mechanically alloyed by high-energy ball milling in a planetary mill in an air atmosphere. Comparing results from electron energy chemical analysis and X-ray diffraction analysis it was found that hardness of the alloy corresponds to the chemical and phase homogeneity of the composite powder. After 24 hours of milling, the single phase homogenous composite powder had the Vickers hardness of about 750 HV1 and coercivity about 40 $A \cdot m^{-1}$. Further alloying led to powder grain size and crystallinite size refinement. This process was accompanied by an increase in the defects amount and thus with increasing of coercivity, up to 120 $A \cdot m^{-1}$ after alloying for 120 hours.

Keywords: soft magnetic materials, Permalloy, mechanical alloying

INTRODUCTION

Soft magnetic materials (SMM) are used extensively in power electronic circuits, as voltage and current transformers, saturable reactors, magnetic amplifiers, recording heads, inductor and transformer cores. These magnetic devices may be required to operate at only 50/60 Hz, or at frequencies down to dc or over 1 MHz [1]. The SMM requirements nowadays are not only for high saturation magnetization, low coercivity, high permeability, low core loss and high resistivity, but also for improved thermal characteristic, flexible machine design and a prospect for greatly reduced weight and production cost. The components manufactured from SMM are generally distinguished by a more integrated design as they consist of fewer parts and, besides, SMM allow construction of devices of more complex geometries. Powder metallurgy (PM) technology is a common economic and environmentally-friendly processing technique for mass production of near-net-shape parts [2].

Two basic types of soft magnetic materials are extensively used, depending on the application and its requirements: ferromagnetic materials based on iron and nickel (for lower frequencies < 2 kHz) and ferrimagnetic materials based on ceramic oxides of metals (for frequencies from a few kilohertz to well over 80 MHz) [3]. Permalloys are Ni-Fe alloys characterized by high amount of Ni, between 70 at.% and 90 at.%, having ordered FCC crystallographic structure of $FeNi_3$ type. Many kinds of Ni-Fe alloys of Permalloy type were developed, doped by a wide variety of atoms to affect the anisotropy and magnetization of the alloy [4].

Hynek Hadraba, Roman Husák, Pavla Roupcová, CEITEC IPM, Institute of Physics of Materials, v.v.i.; Academy of Sciences of the Czech Republic; Brno; Czech Republic
Magdaléna Strečková, Institute of Materials Research, Slovak Academy of Sciences, Watsonova 47, 043 53 Košice, Slovak Republic

Mechanical alloying is a composite powder processing technique involving repeated welding and fracturing of powder particles in high-energy ball mill at low temperature. The process was developed in 1970´s for preparation of structural steel containing high-temperature stable oxide precipitates [5,6] and can be used for overcoming limitations of solid solution forming in liquid state by a conventional metallurgical process [7].

The aim of this work was focused on preparation of Permalloy type SMM by a mechanical alloying preparation route: to prepare very fine powder particles - to be characterized by XRD, SEM and EDX analysis. One-axis compaction of the prepared Permalloy type powder was employed for the preparation of the final samples, used for further mechanical and magnetic tests. The base magnetic measurements confirmed that sufficiently low coercive force and high permeability enables one to tailor a new prospective class of soft magnetic materials with several possible high-frequency applications.

EXPERIMENTAL

The soft magnetic material of Permalloy type was prepared by mechanical alloying process from atomic and pre-alloyed powders summarized in Table 1.

Tab.1. Powders used for 80Ni-14.7Fe-4.4Mo-0.5Mn-0.3Si Permalloy composite powder preparation.

	Ni	Fe	Fe	Mo	Mn	85Fe15Si
Producer	GTV	GTV	Höganas	Sigma-Aldrich	Sigma-Aldrich	PK Chemie
Purity	99%	99%	99.8%	99.99%	99%	-
Particle size [μm]	50	50	100	10	50	10-50
Hardness HV0.1	640	610	125	1530	200	800
Theoretical density [$g \cdot cm^{-3}$]	8,908	7,874	7,874	10,28	7,21	5,802
Permalloy GTV [wt.%]	80 wt.%	13 wt.%		4.4 wt.%	0.5 wt.%	2 wt.%
Permalloy ABC [wt.%]	80 wt.%		13 wt.%	4.4 wt.%	0.5 wt.%	2 wt.%

The blend of powders having nominal composition 80Ni-14.7Fe-4.4Mo-0.5Mn-0.3Si (in wt.%) was mechanically alloyed using planetary ball mill Pulverisette P-6 (Fritsch). The milling was carried out in a hardened steel vial with steel balls in air . The ball-to-powder ratio was 15:1 and rotational speed of main disc was 350 rpm. The microstructure of the composite powders was observed by means of scanning electron microscopy (SEM, JEOL 6460 with Oxford Instruments INCA Energy EDX). Phase composition was determined by X-ray powder diffraction with CoKα radiation (XRD, X´Pert Pro PanAnalytical). Vickers hardness of the powders was measured on an instrumented hardness tester (ZHU Zwick/Roell) under load 0.1 kg according to the EN ISO 6507 standard. The AC hysteresis loops were measured using MATS-2010SA.

Two kinds of dense bulk samples were prepared. For coercivity measurement cylindrical shape samples with dimensions of 10 x 3 mm (d x h) were prepared by cold uniaxial compaction under a load of 800 MPa, followed by pressureless sintering at 1200°C in an Ar atmosphere. Magnetic measurements were conducted on toroid-shaped samples

with outer diameter of 24 mm, inner diameter of 17 mm and height of 2 mm. The sintered samples were prepared both from as-prepared composite powder and from powder pre-annealed at 600°C in an Ar atmosphere.

RESULTS AND DISCUSSION

Evolution of particle size of Permalloy GTV composite powder during mechanical alloying is given in the Fig.1. Particle size was evaluated by means of SEM image analysis using ImageJ software.

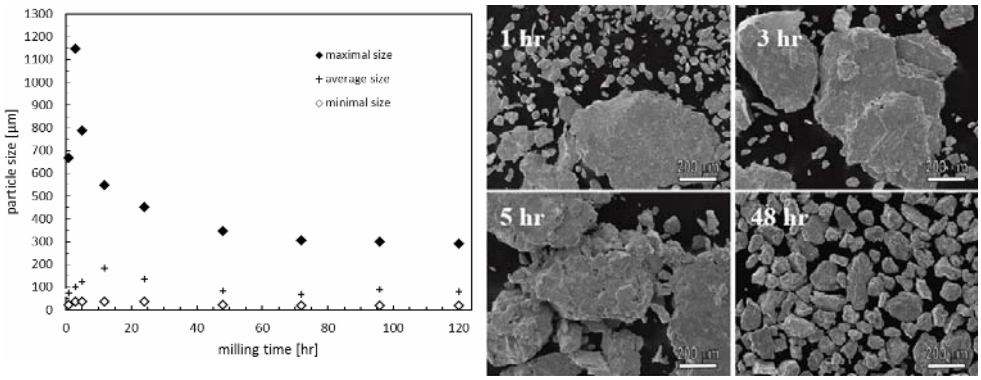

Fig.1. Dependence of particle size on milling time (left) and evolution of particle shape (right) of Permalloy ABC composite powder during milling.

During mechanical alloying the powder particles are cold welded together under impact and shear forces between milling balls and between milling balls and wall of milling vial. Process of particles welding culminates during first 3 hours of alloying when powder contains broad particle size distribution of initial powders having particle sizes from 10 to 100 µm (see Table 1) and composite particles having size of hundreds of µm.

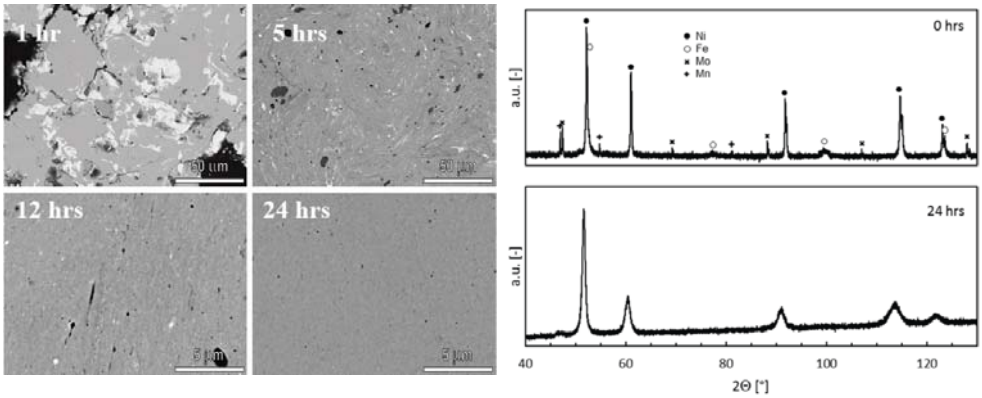

Fig.2. Evolution of microstructure of the composite powder during milling (left) and XRD patterns of Permalloy ABC composite powder before and after 24 hrs of milling (right).

During milling microstructural homogeneity of the composite powder increases (see Fig.2). X-ray patterns confirmed decreasing amount of minority phases (Fe, Mo and Mn), drop in crystalline size and expansion of crystalline lattice of solid solution (Ni-phase)

during milling. After 24 hours of milling the powder consists of one phase having crystallographic structure of the majority phase (Ni). In Figure 3 the Vickers hardness of the powder during milling is plotted.

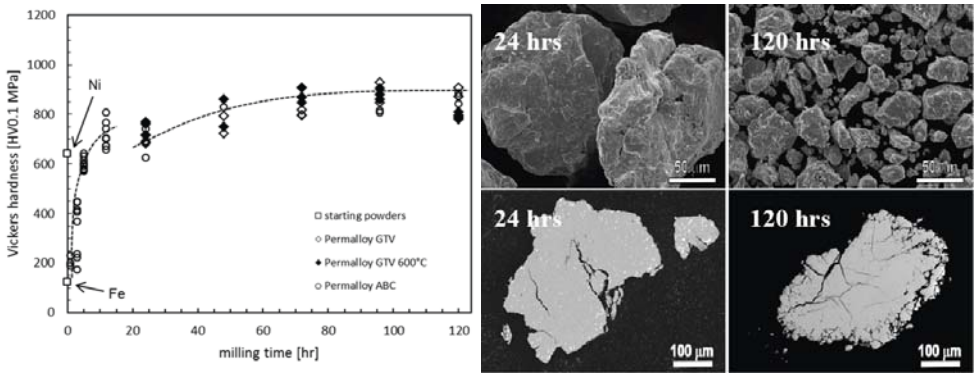

Fig.3. Dependence of composite powders hardness on milling time (left) and change of Permalloy GTV particle shape and macrostructure during milling (right).

In the first 24 hours of milling hardness of the powder increases from 200 to 750 HV0.1. At this moment the powder consists of homogenous single phase solid solution. Further milling leads to powder refinement due to inducing of massive plastic deformation to the particles. In Figure 3 it is visible that after 120 hrs of milling plastic deformation produces massive cracking of particles and their disintegration. The Vickers hardness evolution during milling is reflected in the densities of the specimens prepared from composite powders.

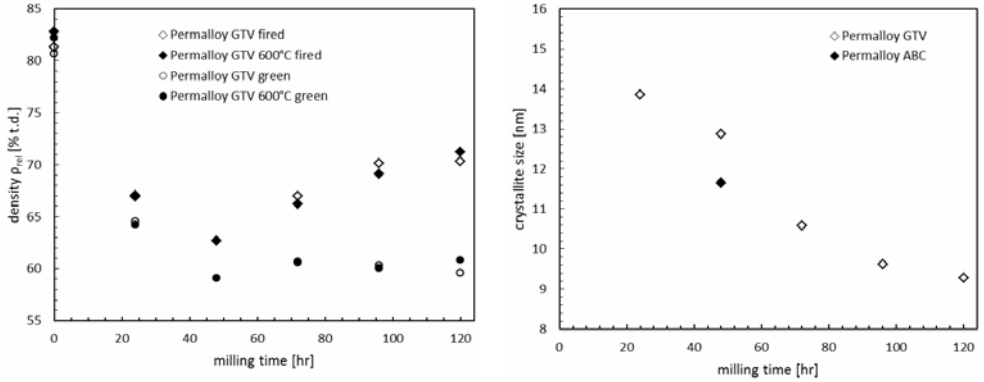

Fig.4. Dependence of green and fired density of Permalloy monoliths on milling time (left) and change of crystallite size of composite powders during milling (right).

In Figure 4 the dependence of the green and sintered relative densities of the specimens are given. The sintered densities were evaluated from dimensional measurements and weighing of the sintered bodies and related to the theoretical density (t.d.) of the composite powder. Theoretical density ρ_{theor} [g·cm^{-3}] of the composite powder was evaluated as 8.72 g·cm^{-3} using equation:

$$\rho_{theor} = \frac{1}{\sum_i \frac{wt.\%_i}{\rho_{theor-1}}} \quad (1)$$

where wt.% is content of i^{th} component in composite powder and $\rho_{theor-t}$ [g·cm^{-3}] is theoretical density of i^{th} component.

The green densities were evaluated from sintered densities, because of the very low toughness of as-pressed green bodies preventing their dimensional measurement. Assuming mass conservation law, it can be derived that ratio of starting density (i.e. the green density ρ_{rel-g}) and final density (i.e. the fired density ρ_{rel-f}) of the bulk is proportional to ratio of final volume V and starting volume V_0 of the bulk:

$$\frac{V}{V_0} = \frac{\rho_{rel-g}}{\rho_{rel-f}} \quad (2)$$

Because the final length l_i [m] of bulk after sintering is given by:

$$l\_i = l_{i0} \cdot (1+\varepsilon_i), \text{ for } i=x,y,z, \quad (3)$$

where l_{i0} [m] is green length of bulk in *x*, *y* and *z* direction and ε_t [%] is relative shrinkage of the bulk. The ratio of final volume *V* [m^3] and starting volume V_0 [m^3] of deposit can be then written in terms of relative shrinkage:

$$\frac{V}{V_0} = (1+\varepsilon_x).(1+\varepsilon_y).(1+\varepsilon_z) \quad (4)$$

If we assume uniform shrinkage of the bulks during sintering (i.e. $\varepsilon_x = \varepsilon_y = \varepsilon_z$)), the fired densities can be recalculated to green densities assuming Eq.(2) and Eq.(4). The Dependence of green densities of Permalloy GTV powders on milling time is given in the Fig. 4. The green density of just blended powder was about 81% t.d. and increased only to 83% t.d. during sintering. For milled powders the green and sintered density of the specimens decreased dramatically as a consequence of hardness increasing (compare Fig.3 and Fig.4). Samples prepared from powders milled for 48 hrs and more had similar green densities: about 60% t.d. This was caused predominantly by decreasing of particle size distribution spread and stabilization of average particle size with milling time (compare Fig.1 and Fig.4). On the other hand, the sintered densities of specimens prepared from powder milled for 48 hrs and more exhibited increase of sintered densities with milling time. This observation can be related to the rapid decrease of crystallite size of composite powder with milling time. The massive plastic deformation caused an increase in the dislocation density and separation of the crystallites into small parts. Stored deformation energy in the form of dislocations and vacancies can then lead to an increase in sintering shrinkage, due to acceleration of diffusion. The Vickers hardness of sintered Permalloy GTV samples in Fig.5 reflected thesintered density of the samples (compare left dependencies given in Fig.4 and Fig.5). As is evident from Fig.5, Vickers hardness of sintered Permalloy GTV samples was proportional to the sintered densities of the samples.

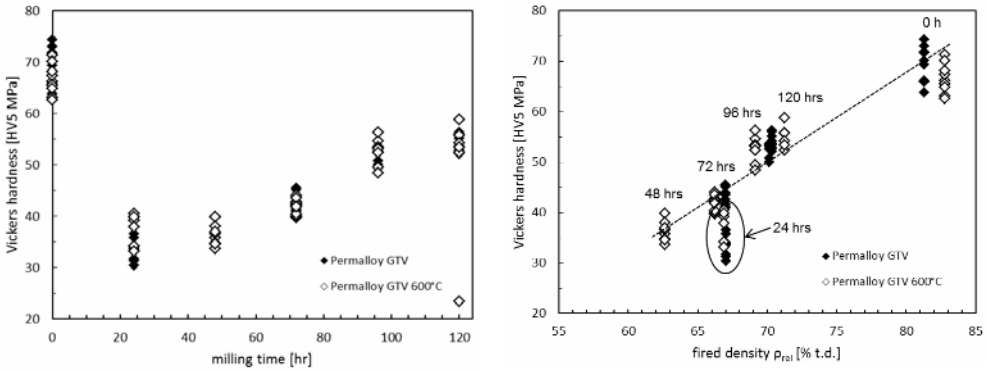

Fig.5. Dependence of hardness of sintered Permalloy monoliths on milling time (left) and on fired relative density (right).

The specimens prepared from powder milled for 24 hours exhibited lower Vickers hardness values, probably due to a wide particle size distribution (see Fig. 1) and thus higher amount of porosity. The dependence of coercivity on milling time is given in the Fig. 6. After 24 hours of milling the coercivity of both Permalloy materials was about 40 $A \cdot m^{-1}$. Further alloying led to increasing coercivity, up to 120 $A \cdot m^{-1}$ and 150 $A \cdot m^{-1}$ for Permalloy GTV and Permalloy ABC, respectively. Physical properties of alloys, and thus of soft magnetic materials, are determined by solid solution ordering. Solid solution prepared by mechanical alloying contains a large number of point and line lattice defects, pores and impurities, resulting in internal stresses. The content of lattice defects affected mainly coercivity of the Permalloys prepared. As a consequence of this, the measured coercivity of Permalloy GTV was proportional to sintered density of the samples (see Fig.6).

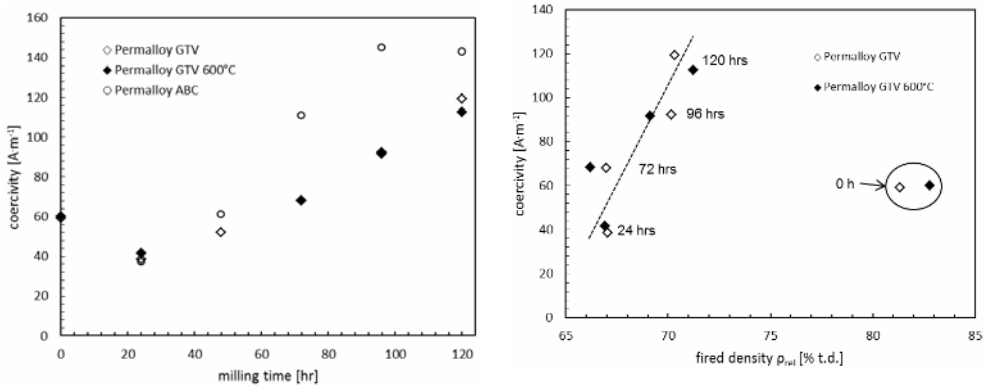

Fig.6. Dependence of coercivity of Permalloy monoliths on milling time (left) and on relative fired density of sintered monoliths (right).

CONCLUSIONS

The 80Ni-14.7Fe-4.4Mo-0.5Mn-0.3Si Permalloy composite powder was successfully prepared from atomic powders by high-energy ball milling in a planetary mill in an air atmosphere and subsequently compacted by uniaxial pressing. The XRD analysis

confirmed that he process of homogenous composite particle formations finished in 12 hours of milling. Alloying is connected with increasing of Vickers hardness of composite particles: hardness of the alloy corresponds to the chemical and phase homogeneity of the composite powder. Further alloying led to powder grain size and crystallinite size refinement. The particle refinement had positive effect by increasing the sintered density of Permalloy material and thus final coercivity. After 24 hours of milling, the Vickers hardness of composite powder was about 750 HV0.1 and coercivity of Permalloy prepared from this powder was about 40 $A \cdot m^{-1}$. Further alloying was accompanied by an increase in the amount of defects and thus with increasing coercivity up to 120 $A \cdot m^{-1}$ after alloying for 120 hours.

Acknowledgement

This work was realized in CEITEC - Central European Institute of Technology with research infrastructure supported by the project CZ.1.05/1.1.00/02.0068 financed from European Regional Development Fund and financially supported by the Czech Science Foundation project No. 14-25246S and Slovak Research and Development Agency under contract no APVV 0222/10.

REFERENCES

[1] Bas, JA., Calero, JA., Dougan, MJ.: J. Magn. Magn. Mater., vol. 254–255, 2003, p. 391
[2] Das, J., Chandra, K., Misra, PS., Sarma, B.: J. Magn. Magn. Mater., vol. 320, 2008, p. 906
[3] Chin – Wen Chen: Magnetism and metallurgy of soft magnetic materials. Amsterdam, New York : North-Holland Publishing Company, 1977
[4] Vernyhora, IV., Tatarenko, VA., Bokoch, SM.: Thermodynamics of f.c.c.-Ni–Fe Alloys in a Static Applied Magnetic Field. SRN Thermodynamics 2012, 2012. 11 p.
[5] Benjamin, JS.: US Patent 3,591,362, 1971
[6] Fischer, JJ.: U.S. Patent 4,075,010, 1978
[7] Shokrollahi, H.: Materials and Design, vol. 30, 2009, p. 3374