

## MAGNETIC PROPERTIES OF A CO BASED AMORPHOUS ALLOY AFTER THERMAL AND MAGNETIC TREATMENTS

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*Sunt prezentate rezultatele obținute pe un eșantion de formă toroidală realizat dintr-un aliaj amorf cu următoarea structură:  $Co_{67}Fe_{4}B_{14.5}Si_{14.5}$  ce a fost supus unui tratament termomagnetic de recoacere în câmp magnetic longitudinal, respectiv transversal. Au fost analizate variațiile pierderilor specifice de energie în funcție de frecvență aplicată (5Hz ÷ 5 MHz) pentru trei valori distincte ale inducției magnetice (5 mT, 10 mT, 20 mT). Separarea pierderilor specifice totale de energie pe cele trei componente (pierderi de energie clasice, pierderi de energie prin histerezis și pierderi de energie în exces) s-a realizat utilizând teoria statistică a lui Bertotti considerând o aproximare liniară pentru variația magnetizației.*

*A toroidal sample of  $Co_{67}Fe_{4}B_{14.5}Si_{14.5}$  amorphous alloy, transversally and longitudinally annealed, was tested to determine the variation of the energy loss as function of frequency in the range of 5 Hz to 5 MHz at a given magnetic flux density (5mT, 10mT, 20mT). The loss separation in the case of amorphous ribbon was made using Bertotti's theory in the approximation of linear magnetization law and arbitrary frequency.*

**Keywords:** amorphous Co based alloy, loss separation, excess losses

### 1. Introduction

The application of amorphous alloys in electrical devices and industrial transformers are increasingly being adopted, helping to solve global warming and energy saving problems. All the industrial applications of amorphous soft alloys are possible because of faster flux reversal, lower magnetic loss and a very easy way to modify their properties [1].

The amorphous alloys can be obtained by rapid solidification method as thin laminations and ribbons. The liquid alloy is placed nearly in contact with a rotating metallic dram at a velocity of 10 ÷ 40 m/s. In this way it ensures a cooling rate of approximately  $10^5$  ÷  $10^6$  °C/s through the glass transition temperature. In

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this state the alloy has the typical viscosity of a solid and, at the same time, the disordered atomic arrangement of a liquid.

The principal composition of amorphous alloys is  $T_{70-80} M_{30-20}$ , where T is one of the transition metals (Fe, Co, Ni) and M it is a combination of metalloids (B, Si, P, C). The radius of the metalloid atoms is much smaller than that of metals and their primary role is to provide the eutectic composition necessary to achieve the amorphous state. Lack of crystalline order does not prevent the formation of ferromagnetic order. The existence of a large-scale magnetic moment is basically unaffected by disorder, although its strength is reduced by the presence of the metalloids. In the case of amorphous alloy the Curie temperature is lower than the corresponding crystalline alloy [2].

The Co-based alloys are ideal materials as cores of inductive components used up to frequencies of 1 MHz in digital telecommunications circuits [2]. These alloys are characterized by vanishing magnetostriction and their magnetic properties can be largely modified by means of field annealing treatments. As mentioned in [4] where it was studied a  $Co_{71}Fe_4B_{15}Si_{10}$  amorphous ribbons that were subjected to a series of field annealing treatments at temperatures  $200 \div 250$  °C. On those samples they induced uniaxial anisotropies, ranging between  $K_u = 100$  J/m<sup>3</sup> (longitudinal, with square hysteresis loop) and  $K_u = -84$  J/m<sup>3</sup> (transverse, with almost linear hysteresis loop). It was observed that the influence of stress anisotropies becomes negligible in the vanishing magnetostriction Co-rich alloys. Accordingly, we expect that  $Co_{67}Fe_4B_{14.5}Si_{14.5}$  material present the lowest energy losses and the highest permeability at all frequencies. Their properties can be modified to specific needs by suitable thermal treatments under a saturating magnetic field. These can induce a large-scale anisotropy,  $K_u$ , as a consequence of localized atomic rearrangements having a definite directional order. Being the only form of anisotropy present in the material,  $K_u$  fully governs coercivity, permeability and loop shapes [2].

In this paper are presented the magnetic properties of  $Co_{67}Fe_4B_{14.5}Si_{14.5}$  amorphous alloy which was characterized using a hysteresis loop measuring setup (Fig. 1). This imposes a prescribed time dependence of the material polarization  $J$  by means of a digitally controlled recursive technique. After a first measurement with the  $e(t)$  waveform, it is obtained an approximate  $J(H)$  relationship. After that an appropriate  $i_H(t)$  function is computed and a new  $e(t)$  function is generated. This process is iterated until the form factor criteria for  $J(t)$  is met [1]. Acquisition of primary current, supplied through a DC-10 MHz NF-HSA4101 type power amplifier, and secondary voltage was made by a 500 MHz TDS 714L oscilloscope

The whole system was driven by software in a VEE environment. In the MHz range, for the minimization of any effect by the stray parameters, it was used very short leads and few-turn bifilar windings.

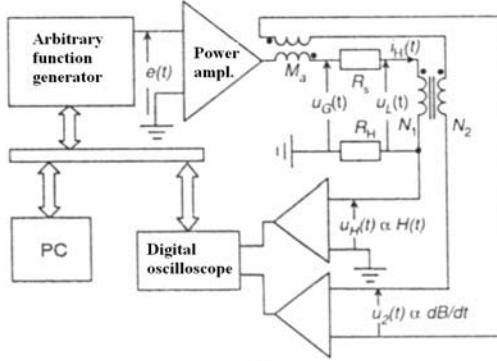


Fig. 1. Block diagram of the hysteresis loop measuring setup

## 2. Experimental results

Two toroidal samples of  $\text{Co}_{67}\text{Fe}_{4}\text{B}_{14.5}\text{Si}_{14.5}$  amorphous alloy, transversally and longitudinally annealed, were tested to determine the variation of the energy loss as function of frequency in the range of 5 Hz to 5 MHz at a given magnetic flux density (5 mT, 10 mT, 20 mT). The physical properties of these samples are given in Table 1.

Table 1

The physical properties and the treatment applied to the sample.

Sample	Sample Characteristics			Applied Treatment
	Density $\delta$ [kg/m <sup>3</sup> ]	Toroidal Section $S_t$ [m <sup>2</sup> ]	Medium Length $L$ [mm]	
1	7730	$0,705 \cdot 10^{-6}$	493,4	-Longitudinally applied magnetic field at 250 °C for one hour
2	7730	$0,613 \cdot 10^{-6}$	504,7	-Transversally applied magnetic field at 250 °C for 15 minutes, -Transversally applied magnetic field at 250 °C for 30 minutes.

For the two samples we obtain the following dependencies for total energy losses (Fig. 2 and Fig. 4) and relative magnetic permeability (Fig. 3 and Fig. 5):

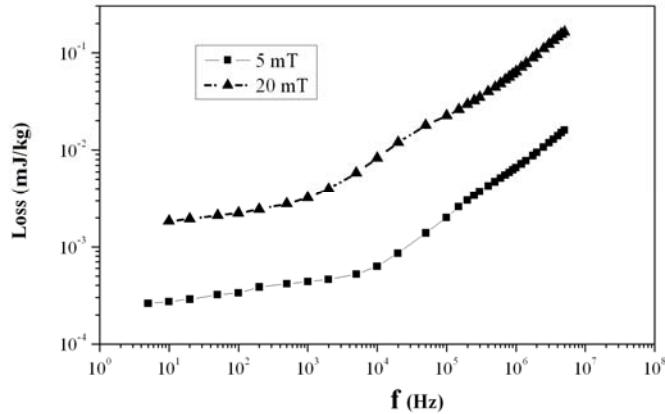


Fig. 2. Losses variation with frequency for  $\text{Co}_{67}$  amorphous ribbon annealed in longitudinally magnetic field

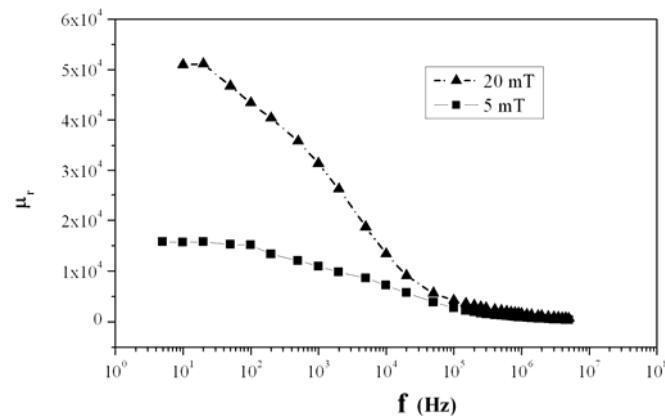


Fig. 3. Magnetic permeability variation with frequency for  $\text{Co}_{67}$  amorphous ribbon annealed in longitudinally magnetic field

Up to  $10^5$  Hz the amorphous alloy treated in transversal magnetic field presents an almost constant relative magnetic permeability which makes this material a good substitute for ferrites cores (Fig. 5). In this frequency range can be consider that the relative magnetic permeability is independent of the frequency because the magnetization of the material it is made by rotation of magnetic moments.

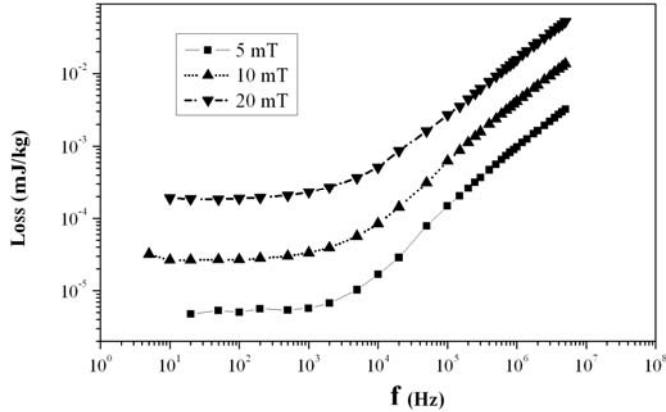


Fig. 4. Energy loss variation with frequency for  $\text{Co}_{67}$  amorphous ribbon annealed in transversal magnetic field

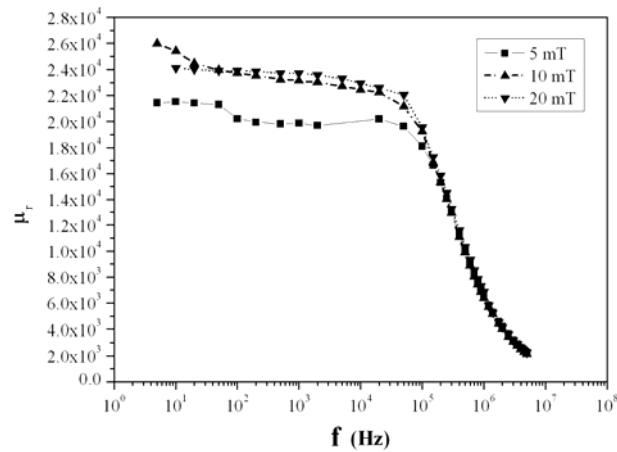


Fig. 5. Magnetic permeability variation with frequency for  $\text{Co}_{67}$  amorphous ribbon annealed in transversal magnetic field

Accordingly to theory and general experiments, we expect that the total loss  $W_{tot}(f)$  presented in Figs. 2 and 4, to increase with frequency as  $f^n$ , with  $n$  ranging from 1.5 and 2. This loss behavior appears to be related with domain walls displacements during the magnetization process of the longitudinally annealed ribbons [4, 5]. With a dominant transverse induced anisotropy the rotation of the magnetic moments is primary magnetization mechanism and we obtain a smaller value of the losses than in the case of longitudinally annealed samples.

At the peak induction  $J_p = 5$  mT the dynamic hysteresis loops are presented (Fig. 6 and Fig. 7) on Rayleigh domain (low magnetic inductions). One

can observe the elliptical shape of the cycles on both cases of thermo-magnetically treatments.

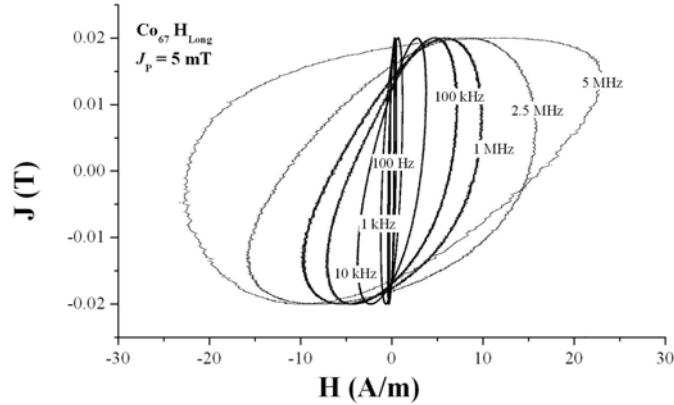


Fig. 6. Evolution of  $J(H)$  dependences with an increasing frequency range in a Co-based amorphous ribbon at a peak polarization  $J_p = 5$  mT on a tape-wound ring samples annealed under longitudinal saturating field

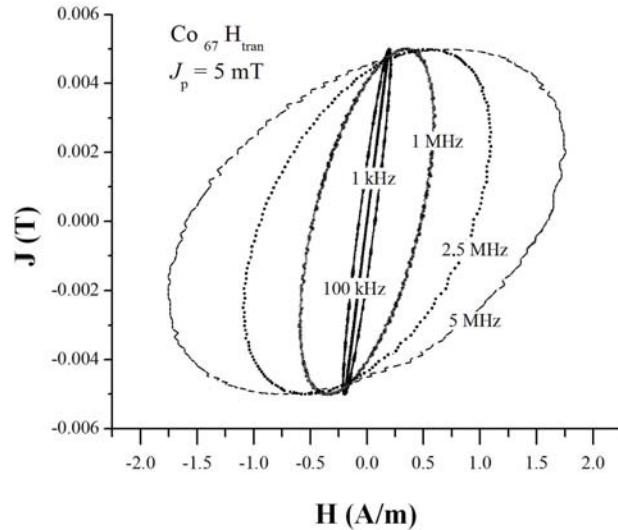


Fig. 7. Evolution of  $J(H)$  dependences with an increasing frequency range in a Co-based amorphous ribbon at a peak polarization  $J_p = 5$  mT on a tape-wound ring samples annealed under transversal saturating field

From the analysis of Fig. 6 and Fig. 7 can be observe that the fundamental consequence of the viscous effects associated with long-range eddy currents is the increase of the energy dissipated in any cycle, as manifest in the observed

broadening of the hysteresis loop with increasing magnetizing frequency. Similar results were obtained on the Co<sub>71</sub>Fe<sub>4</sub>B<sub>15</sub>Si<sub>10</sub> amorphous ribbon [2, 4]. The dynamic cycles are elliptical, with shapes strongly dependent on frequency, peak induction and material microstructure.

### 3. Analysis and interpretation of the experimental results

The total loss is decomposed into the sum of hysteresis, classical and excess loss components. This separation permit us to individually treat loss mechanisms occurring on different space-time scales once at a time, as if they were independent of each other. Hysteresis losses are the consequence of the fact that on the microscopic scale the magnetization process proceeds through sudden jumps of the magnetic domain walls that are unpinned from defects and other obstacles by the pressure of the external field. The local eddy currents induced by the induction change accompanying the wall jump dissipate a finite amount of energy through the Joule effect. The sum of all jumps gives the hysteresis loss associated with the jump sequence. The energy hysteresis loss is independent of frequency [6]. The classical loss is the loss calculated from Maxwell equations for a perfectly homogeneous conducting medium, that is, with no structural inhomogeneities and no magnetic domains [7]. The direct consequence of the magnetic domains walls movement is the excess loses which are very difficult to determine, because of the great variety of domain structures.

The energy loss separation was made for high frequencies [2] using for the calculation of classical energy loss,  $W_{cl}$ , the following formula [3]:

$$W_{cl} = \frac{\pi}{2} \frac{\gamma B_{max}^2}{\mu \delta} \frac{\sinh \gamma - \sin \gamma}{\cosh \gamma - \cos \gamma}, \quad (1)$$

where:  $B_{max}$  – the peak magnetic flux density;  $\mu$  - magnetic permeability;  $\delta$  - material density;  $\gamma = \sqrt{\pi \sigma \mu d^2 f}$  - dimensionless parameter;  $d$  – thickness of the ribbon ( $d = 11 \mu\text{m}$ );  $\sigma$  - electric conductivity ( $\sigma = 709.2 \text{ kS/m}$ ). Further, we calculated the sum of excess energy losses and hysteresis energy losses,  $W_{diff}$ , as follow:

$$W_{diff} = W_{exc} + W_h = W_{tot} - W_{cl}, \quad (2)$$

where  $W_{tot}$  – total energy losses,  $W_h$  – hysteresis losses and  $W_{exc}$  – excess losses.

Making a graphical extrapolation of  $W_{diff}$  to zero, there was determined the  $W_h$  and after the  $W_{exc}$ . The results are presented in the following figures.

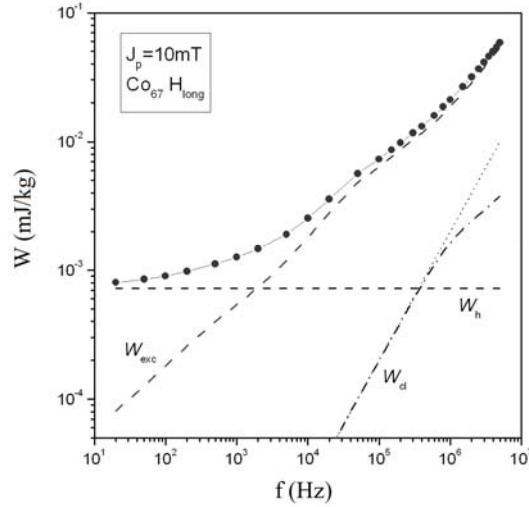


Fig. 8. Energy loss separation as function of frequency for  $J_p = 10$  mT in case of longitudinally treated sample

Fig. 8 and Fig. 10 puts in evidence that, with a well-defined longitudinal domain structure [4], the loss at medium-high frequencies is almost exclusively due to excess loss component  $W_{exc}$ .

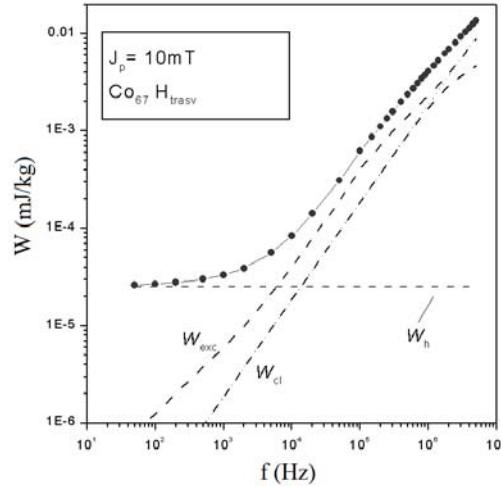


Fig. 9. Energy loss separation as function of frequency for  $J_p = 10$  mT in case of transversally treated sample

Introduced through transverse anisotropy, the rotational processes determine the reduction of the excess loss component  $W_{exc}$  (Fig. 9 and Fig. 11). Similar behaviors were obtained in [2, 4].

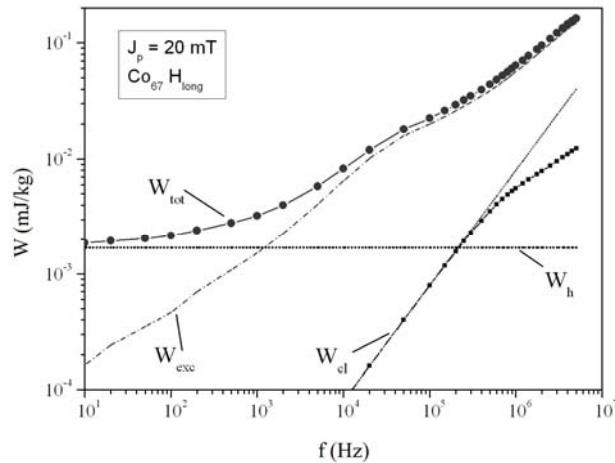


Fig. 10. Energy loss separation as function of frequency for  $J_p = 20$  mT for longitudinally treated sample

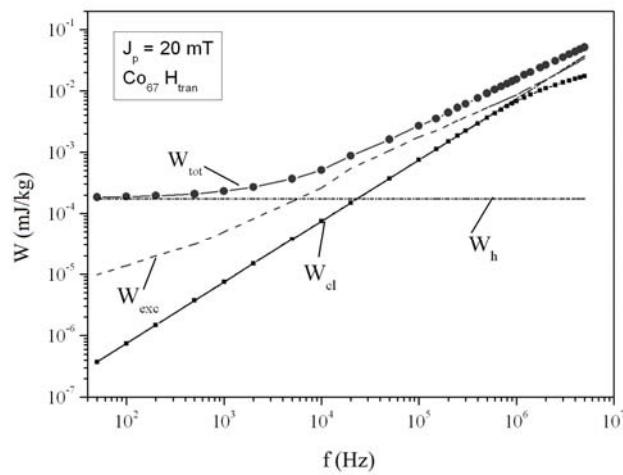


Fig. 11. Energy loss separation as function of frequency for  $J_p = 20$  mT for transversally treated sample

In Co based alloys, fluctuations of local exchange and induced anisotropy should be taken into account, but often, as in all cases where magnetostriction is not negligible, magnetoelastic coupling to randomly distributed, internal stresses dominates and influence the hysteresis losses  $W_h$  [7]. The classical loss  $W_{cl}$ , being related to the large scale eddy currents, is not affected by the specific magnetization mechanism and can be calculated according to standard formulas [5].

#### 4. Statistical interpretation of excess losses

The characterization of the material under controlled conditions can only be done in specialized laboratories. A very large variety of practically encountered waveforms can require a very large database for any specific material, which, in combination with the relatively poor information that can be obtained from the manufacturers datasheets, can determine the prediction of the material energy losses to be very difficult. The model for energy loss prediction based on the statistical theory of losses [8] can be used for soft magnetic alloys if the working frequency is sufficiently low to ensure uniform flux penetration. The model relies on the physically based idea of loss separation, where the total average energy loss  $W(f)$  at a given frequency  $f$  is expressed as the sum of the hysteresis  $W_h$ , classical  $W_{cl}(f)$  and excess  $W_{exc}(f)$  components

$$W(f) = W_h + W_{cl}(f) + W_{exc}(f) \quad (3)$$

where  $W_h$  is considered as a frequency-independent quantity.

The statistical model leads to the conclusion that the large-scale behavior of magnetic domains can be described in terms of the dynamics of  $\tilde{n}$  statistically independent magnetic objects (MO), each corresponding to a group of neighboring interacting domain walls and reduces the loss problem to the investigation of the main physical properties of  $\tilde{n}$  as a function of measurement frequency, peak polarization and material microstructure. This theory gives a natural interpretation of the general fact that the dynamic loss per cycle is a non linear function of measurement frequency. Finally the theory simplifies the dependence of excess energy losses on both peak polarization and measurement frequency to a common mechanism, the competition between the applied field and highly inhomogeneous internal counterfields governing the dynamics of the single magnetic object [9].

The physical mechanism that generates excess losses in soft magnetic materials is based on the competition among the external magnetic field, applied uniformly in the sample, and the local counterfields, highly inhomogeneous, determined by the eddy currents and microstructural interactions. For the

statistical interpretation of losses it is required to apply an external field with the frequency in the range of 0 Hz to 10 kHz. This implies for the calculation of classical losses the following formula:

$$W_{cl} = \frac{\pi^2}{6} \frac{\sigma J_p^2 d^2}{\tau} f = 1.645 \frac{\sigma}{\tau} (J_p d)^2 f, \quad (4)$$

where  $\sigma$  and  $\tau$  are the electrical conductivity and, respectively the density of the material,  $d$  is the thickness of the sample and  $J_p$  is the peak polarization.

For each value of the average polarization rate  $\dot{J} = 4J_p f_{max}$ , the magnetization process in a given cross section of the sample can be described in terms of  $\tilde{n}$  simultaneously active magnetic objects. The dynamic behavior of a single (MO) is controlled by an equation of the form  $H_{exc} \propto \dot{\Phi}$ , where  $H_{exc} = p_{exc} / \dot{J}$  is the excess dynamic field acting on the (MO),  $\dot{\Phi}$  is the magnetic flux rate of change correspondingly provided by the (MO) and the proportionality constant is determined by the damping effect of eddy currents [10, 11, 12]. When there are  $\tilde{n}$  simultaneously active MO's,  $\dot{\Phi}$  must be, on the average, a fraction  $1/\tilde{n}$  of the total flux rate  $S\dot{J}$  imposed to the sample ( $S$  is the cross-sectional area of the probe, so that relationship of the form  $H_{exc} \propto 1/\tilde{n}$  is expected [10], which actually turns out to be:

$$H_{exc} = \frac{p_{exc}}{\dot{J}} = \frac{p_{exc}}{4J_p f} = \frac{H_w}{\tilde{n}} \quad (5)$$

where

$$H_w = 4\sigma G^{(w)} S J_p f \quad (6)$$

and the value of the dimensionless coefficient  $G^{(w)} = \frac{4}{\pi^3} \sum_k \frac{1}{(2k+1)^3} = 0.1356$ .

The fundamental property of the quantity  $\tilde{n}$  appearing in (5) is that it is expected to be a function  $\tilde{n}(H_{exc}; \{P\})$  of the excess field  $H_{exc}$  and of set  $\{P\}$  of parameters characterizing the microstructure and domain structure of different materials. The material  $\text{Co}_{67}\text{Fe}_{4}\text{B}_{14.5}\text{Si}_{14.5}$  treated under longitudinal magnetic field obeys the simple linear law:

$$\tilde{n}(H_{exc}, \{P\}) = \tilde{n}_0 + \frac{H_{exc}}{V_0} \quad (7)$$

where the microstructural information is now carried by  $\tilde{n}_0$ , which represents the limiting number of simultaneously active MO's when  $f \rightarrow 0$  and by the magnetic field  $V_0$ . The phenomenological parameters can be determined by a linear interpolation of the graphical dependence  $\tilde{n}(H_{exc})$ . For the calculus of the excess losses it is used the following formula:

$$W_{exc} = 2J_p(\sqrt{4V_0H_w + (\tilde{n}_0V_0)^2} - \tilde{n}_0V_0)\frac{1000}{\tau}. \quad (8)$$

The energy hysteresis loss was determined through zero extrapolation of the dependence  $W - W_{cl}(f^{1/2})$  which is equal to  $W_h = 0.0017$  mJ/kg.

In the case of  $\text{Co}_{67}\text{Fe}_{4}\text{B}_{14.5}\text{Si}_{14.5}$  the values of the two phenomenological parameters are presented in the Fig. 12. Knowing  $\tilde{n}_0$  and  $V_0$  permits to predict the behavior of  $W_{exc}$  (Fig. 13) through (8).

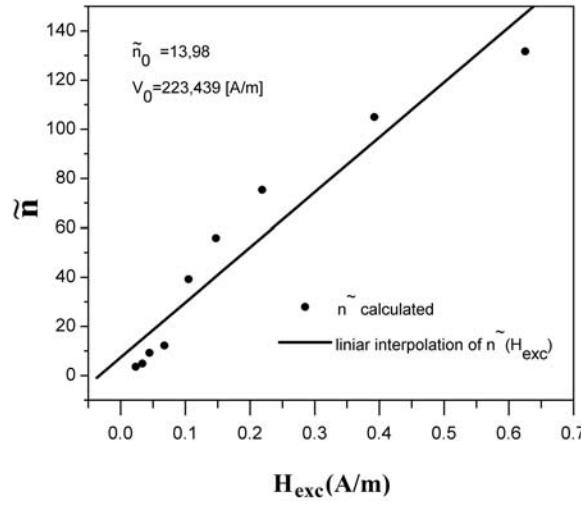


Fig. 12. Representation of the variation of  $\tilde{n}$  versus the dynamic field  $H_{exc}$

The theoretical interpretation of the experimental results on energy excess losses in soft magnetic materials provides a promising convenient tool to look into the connection between dynamic losses and microstructure of samples.

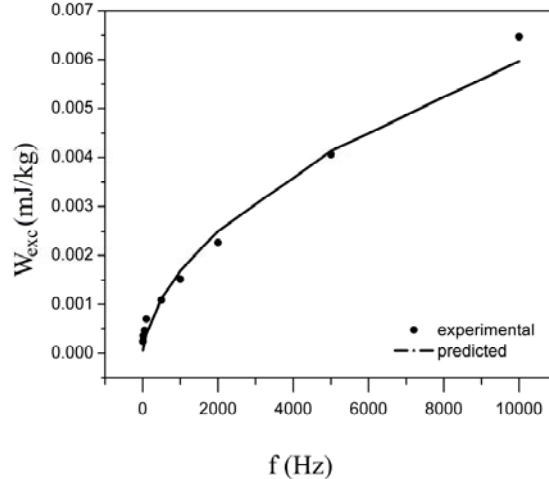


Fig. 13. Comparison between the figures of the excess losses versus frequency in the case of experimental data and predicted values with statistical model

## 5. Conclusions

The alloys that are made most of it of Co are ideal as cores of inductive components to be used up to frequencies of the order of 1 MHz, found in the switched-mode power supplies and in digital telecommunication circuits. Reduce magnetic saturation value is not a disadvantage because, in order to limit core heating, the working induction is always kept small. The loss figure is the fundamental quality parameter used to monitor material production and to select the materials best suited to particular applications [13].

In the case of  $\text{Co}_{67}\text{Fe}_{4}\text{B}_{14.5}\text{Si}_{14.5}$  amorphous alloy the total losses increase with the frequency from  $10^{-3}$  to  $10^{-1}$  and the magnetic permeability decreases with frequency. For the transversally treated alloy can be observed a smaller value of the total loss than in the case of the longitudinally treated alloy, which make this sample very good in the case of applying a longitudinally external field.

The statistical model is a very accurate method for the prediction of the excess energy losses in soft magnetic alloys. The theoretical interpretation of the experimental results on power losses in soft materials has shown that the function  $\tilde{n}(H_{\text{exc}}, \{P\})$  provides a promising tool to look into the connection between dynamic losses and microstructure of soft materials.

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