Rev. Roum. Sci. Techn.– Électrotechn. et Énerg.
Vol. **63**, *1*, pp. 11–14, Bucarest, 2018

Dedicated to the memory of Academician Andrei Ţugulea

HYSTERESIS MODELING ACCURACY FOR SOFT MAGNETIC NANOPOWDERS

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Key words: Hysteresis, Preisach model, Magnetic nanoparticles, Nanopowder, Core-shell nanoparticle, Numerical accuracy.

The paper investigates the experimental and numerical problems related to the macroscopic hysteresis modelling of the soft magnetic core-shell nanoparticles made by $FeNi_3$ nanoparticles, encapsulated in alumina (Al_2O_3) shells. The well-known classical Preisach model was improved by using a Lagrange polynomial interpolation of the numerical Everett integrals and a precise management of the Preisach history line. The numerical tests show an adequate accuracy of the proposed model – error less than 3.2 % for minor hysteresis cycles, even some perturbations occur in the measured data.

1. INTRODUCTION

The magnetic components occupy a large part of electromagnetic conversion systems, but recent researches could improve the numerical simulation of coils [1], the magnetic characteristic measurement [2], the FeSi sheet processing [3] or the losses control for non-sinusoidal regimes [4]. Soft magnetic composites are widely used for electrical machines, transformer yokes or coil cores, the primary material being a magnetic powder. The magnetic properties of nanoparticles (coercivity, saturation magnetization, relaxation time etc.) can be tuned by their composition, size, shape and core-shell structure [5]. The nanopowder experimental characterization is more difficult than for a bulk sample [6]. The coating of the soft magnetic metallic nanoparticles with a layer of insulating inorganic and nonmagnetic material (e.g. Al_2O_3 or SiO_2) generates a core-shell structure [7, 8], which combines the magnetic properties with increased performances at high frequencies, the losses through eddy currents being reduced by the increased electrical resistivity of the particle shell. Our study uses $FeNi_3/Al_2O_3$ nanoparticles that were rarely investigated [9, 10], but have a good potential for electromagnetic shielding, military or space applications [11], even as alternative to the classical FeNi alloy used in power electronics [12].

Our work purpose was the study of classical engineering modelling tools (Preisach hysteresis models) for a hysteretic system (nanopowder) exhibiting a stochastic input that could be modelled as the superposition of a deterministic external input and internal noise [13]. The limits of Preisach modelling are well known, especially for correlated systems [14], but this phenomenological approach is useful for engineer macroscopic simulations [15]. Of course, a detailed testing of the model accuracy must use a standardized environment, as a benchmark problem [16]. Analytical Preisach or Everett functions were tested with good results for bulk soft magnetic materials [17, 18]. Our investigation on the measured magnetization curve for some $FeNi_3/Al_2O_3$ nano-powder samples revealed zones where negative susceptibility occurs. The source of such curves could be the large negative Barkhausen jumps (previously reported for permalloy thin films [19] or Fe-Si monocrystals [20]), a weak measurement accuracy due to the instrument [21] or the particles' movement inside the powder sample. A profounder study could be made with a magneto-optical Kerr technique [22].

This paper proposes an optimized numerical hysteresis model, based on a scalar classical Preisach model, where the identification of the numerical Everett function (integrals of Preisach statistical function) uses a set of first-order reversal curves (FORC). The modeling is appropriate for soft nanopowders, which are more difficult than bulk materials to be characterized and modeled. The model robustness is improved by associating a Lagrange polynomial interpolation to the identified Everett values for the Preisach grid nodes and by an efficient management of the memory line updating. The model was tested for complex magnetization processes, including minor hysteresis cycles.

2. IMPROVED PREISACH HYSTERESIS MODEL

The well-known phenomenological hysteresis model introduced by Preisach is still widely used for macroscopic technical applications, even micro- or nanostructured magnetic materials are involved. A robust numerical implementation can assure an adequate accuracy, avoiding the too complex micromodeling of the magnetic cores involved in technical systems like electric transformers, machines, electromagnetic shields etc. Our numerical model uses the classical scalar Preisach approach, the implementation being optimized to assure a good accuracy and an easy coupling with electromagnetic analysis methods (e.g. finite elements method). The model key features are presented below.

A) A non-uniform grid of the Preisach triangle is used, with a lower density for the saturation zone (Fig. 1). The measured FORCs, which are used for identification, correspond to this grid.

B) The model is defined by a discreet numerical set of the Everett integral values, which are directly computed for the Preisach grid nodes (see Fig. 1):

$$E(i, j) = M(a_i) - M(b_j), \tag{1}$$

where $M(a_i)$ and $M(b_j)$ are the magnetization values measured on FORC no. (j) when the magnetic field increases from $H = b_j$ to $H = a_i$.

C) The history staircase line in Preisach triangle is carefully managed, using the exact reversing points instead the grid adjacent nodes (Fig. 2). For each new reversing point, the associated Everett value is accurately computed by a Lagrange polynomial interpolation between the four known values for the rectangle corners where it is placed.

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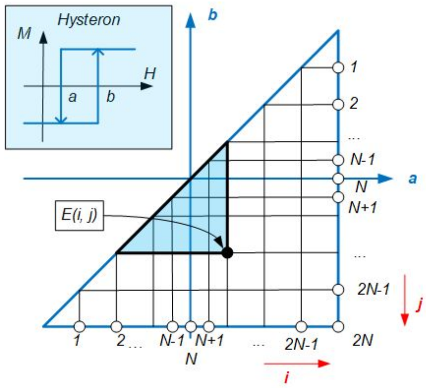

Fig. 1 – Non-uniform grid of Preisach triangle and nodal Everett function value *E* (*i, j*).

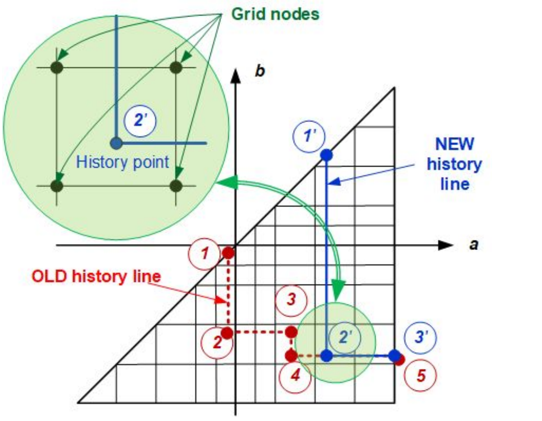

Fig. 2 – History line updating in Preisach triangle from the old state (staircase line 1–2–3–4–5) to the new one (staircase line 1'–2'–3').

The history line updating uses these exact reversing points, not the grid nodes, the accuracy being improved.

The Preisach numerical model is characterized by the set of $E(i, j)$ values in the mesh nodes, which were identified by (1). The history line is updated for each new value H_{new} of the magnetic field. The new magnetization M_{new} is computed from the previous (old) magnetization M_{old} using the Everett values associated to the history line nodes; for example, the increasing of H to the value $a = H_{new}$, presenting in Fig. 2, produces an increasing of M:

$$M_{new} = M_{old} + 2[E(2')–E(4)+E(3)–E(2)], \qquad (2)$$

where $E(k)$ is the Everett integral value associated to the node (k) of the history line.

3. INVESTIGATED SOFT MAGNETIC NANOPOWDER

The studied soft magnetic nanopowder is constituted by individual or aggregates grains of $FeNi_3$ compound, surrounded by Al_2O_3 layers or chains. To prepare by sol-gel method the $FeNi_3/Al_2O_3$ nanocomposites with 5 % Al/(Fe+Ni) molar ratio, appropriate amounts of metallic salts, Fe^{2+} Ni^{2+} and $C_6H_8O_7{\cdot}H_2O$ (1:3:9 molar ratio) were dissolved in ethanol and further addition of 0.16 mol of Al^{3+} salt. The dried gel was annealed for 4 h at 400^{o}C in air and in reducing atmosphere at 800°C for 4 h, similar with [9]. To disperse the nanoparticles homogeneously, a liquid suspension containing the studied $FeNi_3/Al_2O_3$ nanopowder and hexane was for 30 min ultrasonically stirred.

The dispersed nanoparticles have been deposited on the support grid and after drying, the samples microstructure was characterized by transmission electron microscopy (TEM) using a Carl Zeiss® high resolution transmission electron microscope (HR-TEM), model Libra 200FE-HR, at an accelerating voltage of 200 kV. The HR-TEM images of $FeNi_3/Al_2O_3$ nanoparticles presented in Fig. 3 reveal the presence of the atomic planes in $FeNi_3$ crystalline grains, with an interplanar distance of 0.76 nm and a very thin layer of 1.85 nm from amorphous Al_2O_3 (see the enlarged images in Fig. 3). The X-ray diffraction (XRD) pattern in CoK_β radiation for $FeNi_3/Al_2O_3$ nanopowder reveals the $FeNi_3$ compound formation, with cubic crystalline structure and lattice parameter a = 3.5523 Å. The average grain size of the $FeNi_3$ nanocrystallites, calculated by Debye-Scherrer method, was d = 40.1 nm.

A technologically processed nanocomposite has another magnetic property than the original nanopowder and micromagnetic simulations could be useful. Indeed, our core-shell nanoparticles ($FeNi_3/Al_2O_3$) were sintered and a cylinder sample (2.9 mm diameter and height) was measured in similar conditions (room temperature). The nanopowder was compacted at 160 kN/cm^2 and sintered at 1250^{o}C for 1 hour in argon, reaching the density of 7.86 g/cm^3; the measured sintered sample was obtained by electrical discharge machining (EDM). After the demagnetizing correction, the experimental curves show a diminished hysteresis (advantageous for magnetic core losses) and an increased saturation magnetization (Fig. 4). These results prove the material versatility, its properties depending on the technological process.

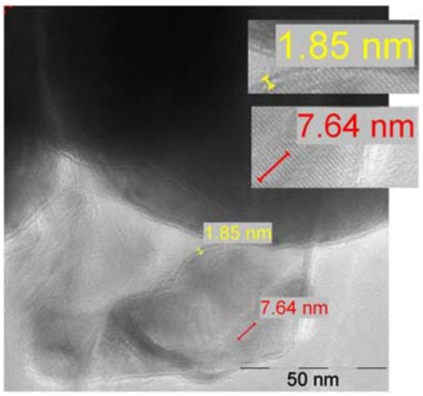

Fig. 3 – HR-TEM images of $FeNi_3/Al_2O_3$ nanoparticles, with highlighting of the Al_2O_3 layer (top) and of the atomic planes in $FeNi_3$ crystalline grains (bottom).

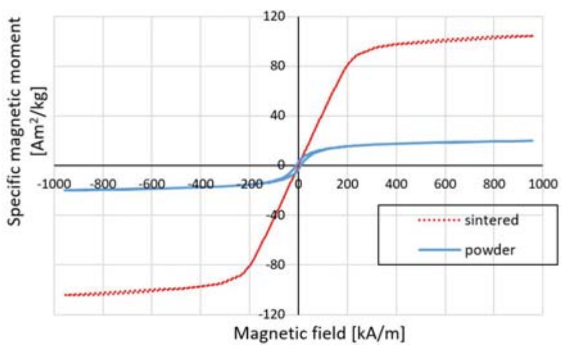

Fig. 4 – Experimental curves for sintered and nanopowder samples of $FeNi_3/Al_2O_3$.

4. IDENTIFICATION OF HYSTERESIS MODEL

The magnetization curve measurement for $FeNi_3/Al_2O_3$ nanopowder samples revealed zones where very small negative or positive susceptibilities occur (Fig. 5), probably due to the strong magnetic interactions between the nanoparticles, clusters and their shells or to the instrument noise.

The experimental FORCs presented in Fig. 5 were measured by a vibrating sample magnetometer (VSM LakeShore 7304) for $FeNi_3/Al_2O_3$ samples. The nanopowder was inserted in the VSM cylindrical holder (3.8 mm diameter), filling a height of 4.2 mm (44.3 mg). The shape of the powder holder and the filling factor influence the magnetization curves [6]. Our challenge was to test if a Preisach numerical model could offer an appropriate accuracy for this behavior associated with a stochastic input (noise).

Preisach model was identified on a non-uniform grid having 8 kA/m field step between (–240 kA/m) and (240 kA/m), and 80 kA/m field step outside this range. The numerical Everett function (integral of Preisach statistical distribution over triangle areas) was directly computed from a set of 78 FORCs and it is presented in Fig. 6.

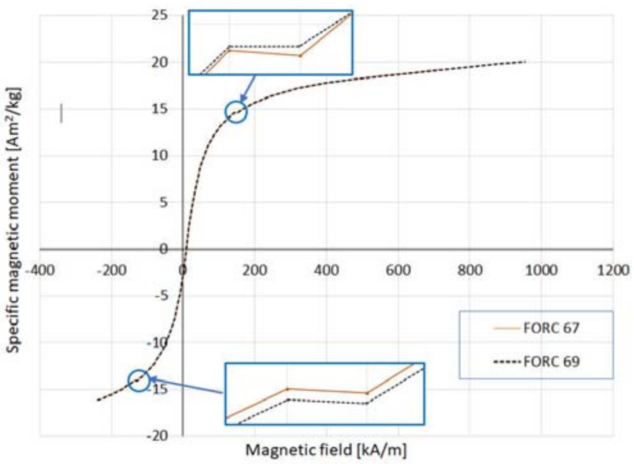

Fig. 5 – Experimental FORCs with perturbations.

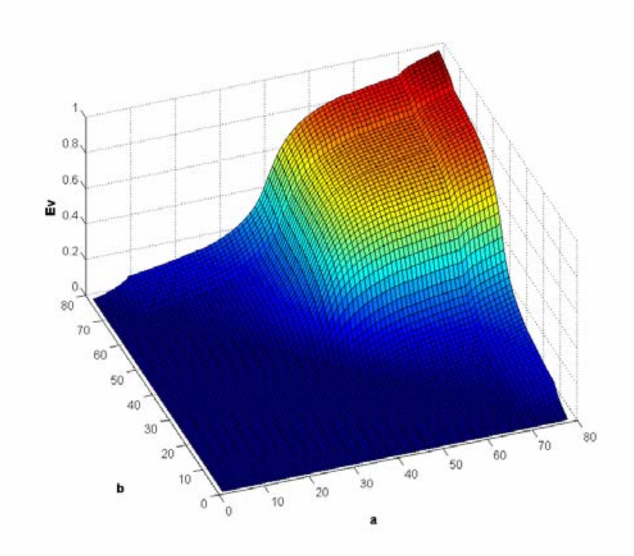

Fig. 6 – Identified values of the Everett function *Ev* in the grid nodes of Preisach plane (*a*, *b*) of the hysterons switching fields.

5. NUMERICAL TESTS AND RESULTS

The model accuracy was tested for ascending and descending branches, involving two symmetrical 2^{nd}-order minor loops [–64 kA/m, 64 kA/m], using the experiment scenario presented in Fig. 7. The obtained numerical results show a good accuracy in respect to the measured ones.

The results presented in Table 1 shows a mean error for the entire profile equal to 1.93 %, with a maximum of 3.19 % for a second-order minor cycle. It must be mentioned that the hysteresis model conserves the perturbed zones, as one can see in Fig. 8.

Table 1

Relative root mean square error (RMSE) between computed and measured magnetization values for the experiment presented in Fig. 7

Zone	Magnetic field evolution [kA/m]	RMSE [%]
1	(+960) → (–64)	1.52
2	(–64) → (+64) → (–64)	3.19
3	(–64) → (–960)	1.77
4	(–960) → (+64)	2.61
5	(+64) → (–64) → (+64)	2.25
6	(+64) → (+960)	0.68
	entire experiment	1.93

The biggest influence on the modelling accuracy comes from the magnetic history management, because the magnetization is incrementally computed and the error could be accumulated. Preisach model depends on the reversal points of the magnetic evolution and their precise updating increases the model accuracy.

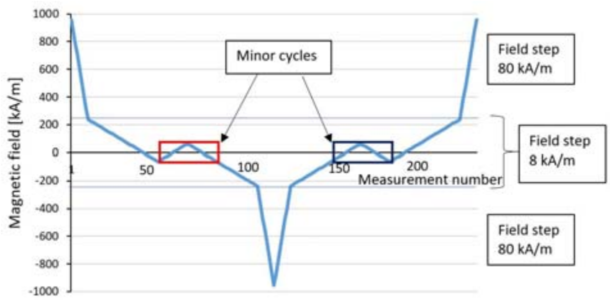

Fig. 7 – Magnetic field evolution with variable field step (8 kA/m and 80 kA/m) for an experiment involving two symmetrical 2^{nd}-order minor loops [–64 kA/m, 64 kA/m].

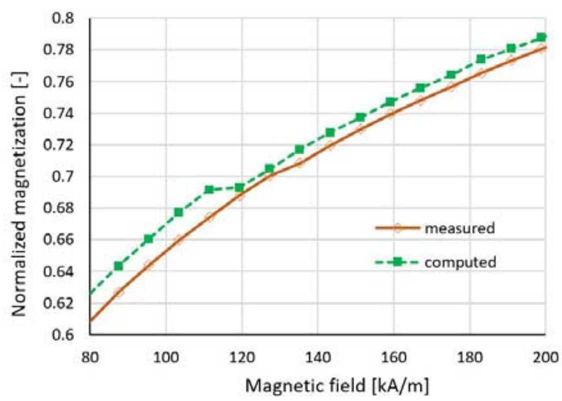

Fig. 8 – Model behavior in zones with negative magnetic susceptibility.

6. CONCLUSIONS

The obtained results prove that the soft magnetic nanopowder locally exhibiting negative susceptibility can be successfully modelled by a numerical classical Preisach model. The numerical errors are minimized by using a direct identification of the Everett integrals starting from FORCs with variable field step.

The hysteresis model accuracy is increased by a precise updating of the reversal points in the material magnetic history, the associated Everett function values being computed by a Lagrange polynomial interpolation between the four identified values for the grid nodes (corners of the corresponding rectangular cell). The numerical tests, including 2nd order minor hysteresis cycles, prove the model accuracy for any zone of the magnetization scenario applied to a core-shell soft magnetic nanocomposite $FeNi_3/Al_2O_3$.

Despite its known limitation, the classical Preisach hysteresis model can be used for the macroscopic computation of nanoparticles systems, as the soft magnetic composite cores, if an optimized numerical processing is assured. A more precise modelling must involve a generalized hysteresis model, including vector or dynamic features, and the reversible magnetization component; the demagnetizing field must also be considered. However, a micromagnetic model of an interacting nanoparticles system could be better, but it is inadequate for an engineering application. Consequently, one still uses phenomenological methods for technical devices made by new and tailored materials, such soft magnetic nanocomposites.

ACKNOWLEDGEMENTS

This work was supported by a grant of the Romanian National Authority for Scientific Research and Innovation, CNCS/CCCDI-UEFISCDI, project No. PN-III-P2-2.1-PED -2016-0451 and by a grant of Romanian Agency for Space, project STAR No. 125/2017, within PNCDI III.

Received on November 24, 2017

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