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MAGNETIC PROPERTIES OF SOME PERMINVAR FERRITES

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Mn-substituted Ni ferrites doped with cobalt and copper ions sintered in some conditions were prepared by a conventional ceramic technique. Structural and magnetic properties of the ferrites were characterized by X-ray diffraction, scanning electron microscopy and high frequency permeability measurements. These studies revealed that the magnetic performances are influenced by the ions content and the microstructure. The contribution of two magnetization mechanisms, domain wall motion and magnetization rotation, to the high frequency complex permeability, is briefly discussed.

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1. Introduction

Due to their high electrical resistivity and low magnetic losses NiZn ferrites are of great commercial importance for their magnetic properties at high frequency (>1 MHz). The recent application, such as multilayer chip inductors as well as suppression of electromagnetic interferences, commonly known as EMI, are currently used in notebook computers, hard disk drivers and other electronic products. Many applications, including some those in the communication, further require that the ferrites exhibit a small change in permeability with frequency or temperature in a range of temperature dictated by the application. The desired characteristic for most applications of these materials are high induction, high permeability, low coercitivity and low power losses.

High resistivity MnNi and MnNiZn ferrites have already been reported [1,2]. Previous studies related the influence of copper [4,5] and cobalt oxides [6,7] to the magnetic properties of NiZn ferrites. It was observed [1] that the saturation magnetization and initial permeability are sensitively controlled by manganese substitution of NiZn ferrites. The losses increase slightly by increasing sintering temperature, while the relaxation frequency decreases as Zn content increase [2]. Copper oxide is useful to obtain low sintering temperature but it is responsible [4-7] for residual losses which affects the dynamical magnetic properties. In order to decrease core losses and achieve good electromagnetic properties a small amount of cobalt oxides was used [7,8]. Divalent cobalt ions on the octahedral sites of the spinel lattice make a large contribution to the magnetocrystalline anisotropy, thereby compensating for the negative anisotropy of the Co free lattice [8]. This result leads to a crossing of the first order magnetocrystalline anisotropy constant (K_1) at some temperature below the Curie temperature, which in turn gives rise to a secondary peak in the permeability-temperature curve. By adjusting the position of the peak trough chemistry and sintering process, one can achieve a temperature range between the two peaks in which the change in permeability is quite small. The same small variation can be observed in the permeability spectra versus frequency (perminvar ferrite). The permeability of perminvar ferrite is constant in a wide range of the frequency. The hysteresis loops are linear in shape at low amplitude of the applied magnetic field. Increasing the applied magnetic field

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beyond the H_p the hysteresis loop becomes open. At high values of the applied magnetic field the hysteresis loop is similar with those of the usually NiZn ferrite. The hysteresis, eddy current and Jordan post action losses are reduced in perminvar ferrite case.

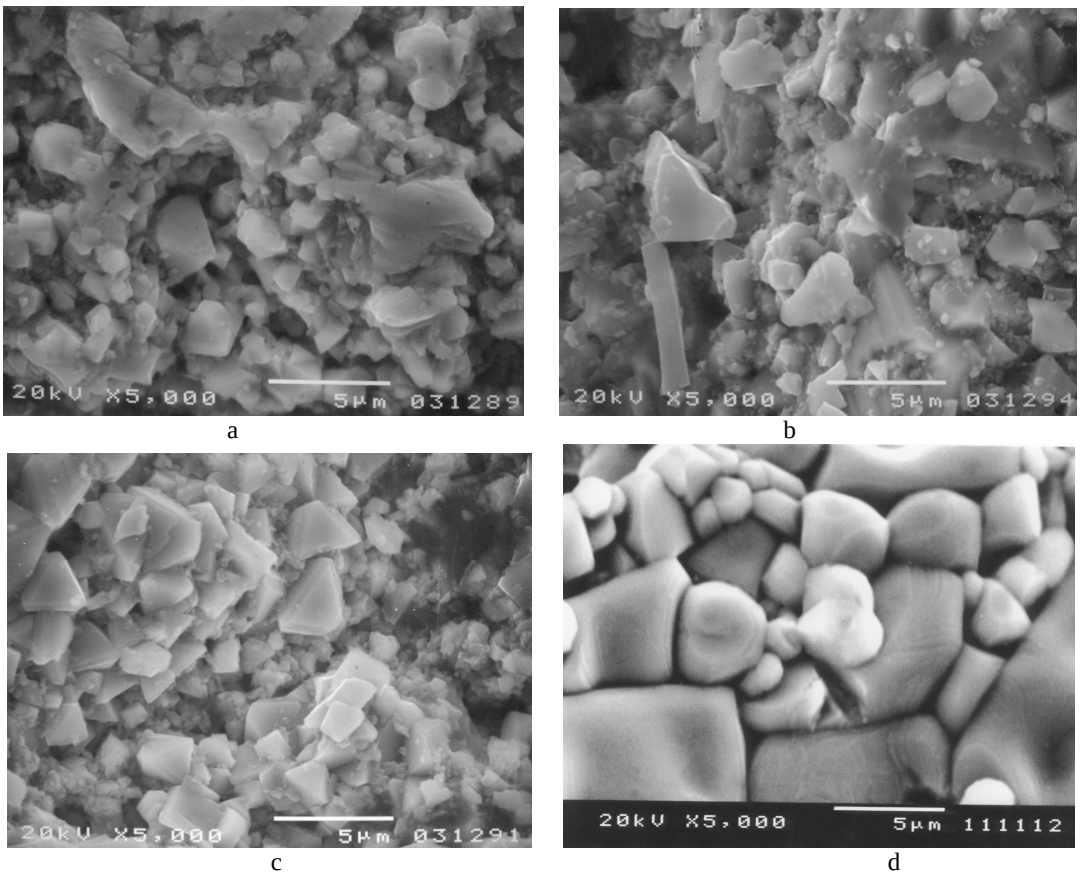
A detailed interpretation of the influence of the manganese, zinc, copper and cobalt ions and parameters of the sintering process were made in [2-4,7]. The purpose of this limited study was to determinate the effect of the different ions content and microstructure on the complex permeability spectra of Mn-substituted NiZn ferrites doped with cobalt and copper oxides.

2. Experimental results

Four categories of samples (labeled D, E, F and G) were considered in our study. The samples are ferrites with iron in excess and doped with cobalt and copper oxides.

The chemical composition of the studied ferrite was as following: D sample $Ni_{0.70}Fe_{2.10}O_4$ added with 1.7wt%MnO and doped with 0.60 wt%CoO; E sample $Ni_{0.35}Zn_{0.28}Fe_{2.20}O_4$ added with 1.7 wt%MnO and doped with 0.30 wt%CoO; F sample $Ni_{0.42}Zn_{0.28}Fe_{2.20}O_4$ added with 1.7 wt%MnO and doped with 0.30 wt%CoO; G sample $Ni_{0.42}Zn_{0.28}Fe_{2.20}O_4$ added with 1.7wt%MnO and doped with 0.30wt%CuO.

Samples were synthesized using the usual ceramic technique [6]. Pure starting powders of αFe_2O_3, NiO, ZnO, CoO, $Ni(OH)_2$.$Ni(CO_3)$ and $MnCO_3$ were mixed in suitable proportions for 20 hours in steel ball-mill using water as the mixing medium. The mixture was dried and calcined at 900^{o}C for 12 hours in air. The presintered powders were milled in water to obtain BET surface 3±0.5m^2/g. The ring cores were pressed at 150 MPa the green density being 2.9±0.2 g/cm^3. All the samples were sintered in air at 1250 ^{o}C for 2h. The cooling rate was 100°C/h.



a b c d

Fig. 1. The microstructure of the samples: (a) – sample D; (b) – sample E; (c) sample F; (d) sample G.

All the samples were structurally characterized using a Philips X'Pert, Cu K_α anode, high-resolution x-ray diffraction system. The θ-2θ scans of the samples demonstrate for samples D, E, F small quantity of remanent αFe_2O_3 phase. Also, it must be noted the sharp peaks states for a high crystallinity of the samples. The characterization of the samples was completed by the EDAX analysis (DXPRIME) that confirmed the different concentration of the Zn, Mn, Co and Cu ions in our samples. In Fig. 2 are featured comparatively the EDAX spectra for sample D and F(Fig. 2). The diagram for sample D states the absence of Zn ions in the composition while the diagram for sample F contains the Zn corresponding peaks.

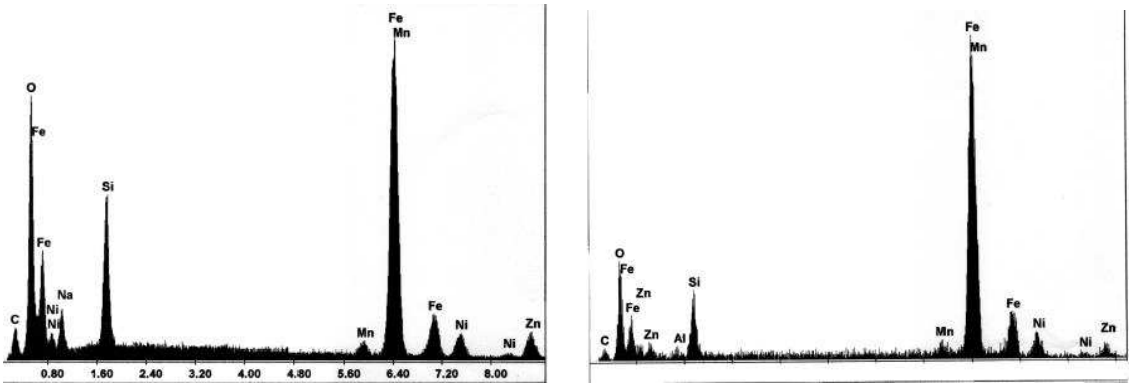


Fig. 2. The EDAX spectra: (a) sample D; (b) sample F.

The microstructure of the samples, presented in Fig. 1, was performed by scanning electron microscopy (SEM), using a JEOL 5410 system. The microstructure of the samples D and F is uniform, the pores being presents at the grain boundaries. In the case of the sample D the average grain size was 1.5 ± 0.2 μm. The sample F had the mean grain size 2.4±0.3μm. Sample E had a non-uniform microstructure with great grain differences, pores and addition segregation at the grain boundaries. The average grain size was 6.7±0.5 μm. Sample G has a compact structure without pores at the grain surface and the average grain size determined by linear interception methods was 8.3±0.6 μm. The microstructure of the sample explains the magnetic properties. The mean dimension of the grains influences the values of the initial permeability. Permeability is greater for the sample G having the greatest mean grain size. The density was determined by archimedic methods. The initial permeability was measured on a core without airgap at room temperature and B<0.1 mT. The resistivity was measured with four probes method and the Curie temperature, T_C, was determined from $\mu = f(T)$ curves. The results are summarized in Table 1. All the complex permeability spectra confirm the microstructure previously observed. The spectra was measured by using the conventional technique based on the determination of the complex impedance of a circuit loaded with a toroid shaped sample (Hewlett Packard 4191B Impedance Analyzer with Option 002 and HP 16454A magnetic material test fixture). The samples D and F feature a perminvar ferrite complex permeability spectrum. The permeability remains almost constant in a large frequency range and the resonance frequency occurs at 200 MHz for sample D ant at 40 MHz for sample F. Increasing the average grain size and the non-uniformity, as in the cases of the samples E and G the resonance frequency moves towards lower values, respectively to 5 MHz and 0.8 MHz.

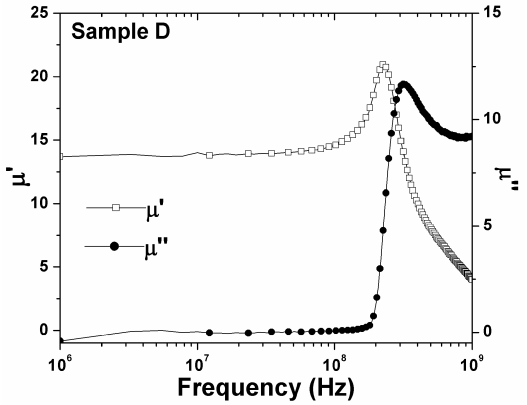


Fig. 3a. Spectra of the complex initial permeability at room temperature for Sample D.

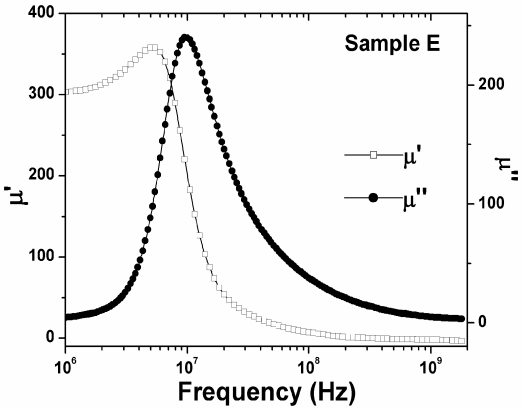


Fig. 3b. Spectra of the complex initial permeability at room temperature for Sample E.

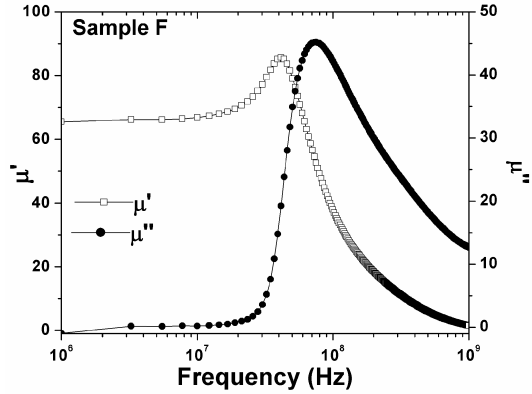


Fig. 3c Spectra of the complex initial permeability permeability at room temperature for Sample F.

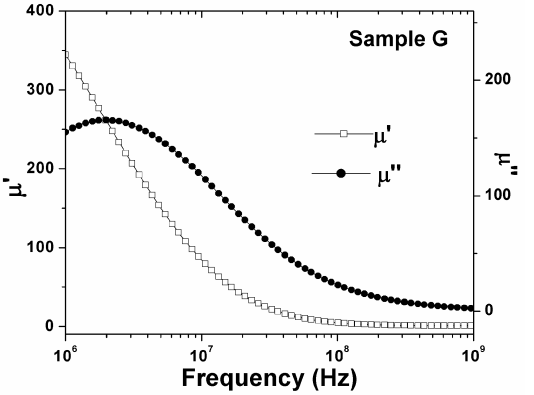


Fig. 3d Spectra of the complex initial at room temperature for Sample G.

The complex permeability is related to two different magnetizing mechanisms: the spin rotational magnetization and the domain wall motion [4,5]. At low frequency leads the magnetization by magnetic wall displacement, while at high frequency rotation component becomes important. The domain wall motion is more sensitive to the microstructure and is controlled by the ferrite grain size. The domain wall motion plays an important role in magnetization processes and losses mechanism in samples E and G characterized by larger grain size.

Table 1.

Samples	D(g/cm^3)	ρ_v (Ω.m)	μ_i	T_C (^{o}C)
D	4.4	10^4	12	450
E	5.0	10^4	300	270
F	4.3	10^4	70	400
G	5.2	10^4	325	416

3. Conclusions

Four categories of ferrites analyzed in this paper demonstrate the sensitivity of the magnetic properties on the composition and the sintering process. One composition without Zn ions illustrating perminvar behavior and few compositions in which the influence of Mn, Co and Cu ions plays an

important role to the microstructure and magnetic properties were studied. The measurements of the density, initial permeability and complex permeability spectra have proved the influence of the microstructure on the magnetization process. Large grain size implies high values of the initial permeability due to the magnetization by displacement of the magnetic domain walls but shifting of the natural resonance to the lower frequency. Small average grain size leads to the processes in which the rotational magnetization plays the determinant role. The value of the permeability and the limiting frequency should represent a compromise for ferrite core user. The sample D could be an example of low permeability ferrite for high frequency while sample G an example of high permeability ferrites for low frequency.

References

[1] K. Singh, T. C. Goel, R. G. Mendiratta, o. P. Thakur and C. Prakash, "Magnetic properties of Mn-substituted NiZn ferrites", J. Appl. Phys. **92**(7), 3872 (2002).
[2] M. I. Rosales, O. A. Valenzuela, R. Valenzuela, IEEE Trans. Magn. **37**(4), 2873 (2001).
[3] M. I. Rosales, R. Valenzuela, Materials Science Forum **302–303**, 411, Trans Tech Publications, Switzerland 1999.
[4] T. Nakamura, J. Magn. Magn. Mat. **168**, 285 (1997).
[5] O. F. Caltun, L. Spinu, A. Stancu, IEEE Trans. Magn. **37**(4), 2353 (2001).
[6] O. F. Caltun, L. Spinu, A. Stancu, L. D. Thung, W. Zhou, J. Magn. Magn. Mat. **242**, 160 (2002).
[7] A. Marais, C. R. Acad. Sci. **248**, 2170 (1959).
[8] R. Lebourgois, J. Ageron, H. Vincent, J. P. Gaune, Proc. Of. ICF'8, Kyoto and Tokyo, 576 (2002).