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Vincent Loyau, G.Y. Wang, Martino Lobue, Frédéric Mazaleyrat

An Analysis of Mn-Zn Ferrite Microstructure by Impedance Spectroscopy, STEM and EDS Characterisations.

V. Loyau[1], G-Y. Wang[2], M. Lo Bue[1], F. Mazaleyrat[1]

[1]*SATIE UMR 8029 CNRS, ENS Cachan, PRES UniverSud, 61 avenue du président Wilson, 94235 Cachan Cedex, France*
[2]*LMPQ UMR 7162, Université PARIS 7, 10 rue Alice Domon et Léonie Duquet, 75205 Paris Cedex 13*

E-mail : vincent.loyau@satie.ens-cachan.fr

Abstract
AC resistivity measurement results on Mn-Zn sintered ferrite were analyzed in the 0.1-500 MHz range. From electrical point of view, the material could be represented by an equivalent circuit of parallel resistance-capacitance cells connected in series corresponding to the contributions from bulk grains in one hand, and grain boundary layers in the other hand. The experimental resistivity curves were fitted with the model. The as obtained parameters give information on dielectric properties and conductivity of both bulk grains and boundary layers. For the studied material, it appears that the resistivity at low frequencies is increased 27 times due to the boundary layers effects.
STEM (Scanning Transmission Electron Microscopy) and EDS (Energy Dispersion Spectrometry) characterization where performed in order to detect impurities at a grain boundary layer which can explain those wide differences between bulk grains and boundary layers electrical properties. It appears that the two components have close chemical compositions, but some calcium impurities segregate at the boundary which increases dramatically the resistivity of these layers.
Furthermore, the bulk grains show relative permittivity around 350 at low frequency which is much smaller than the one measured for the whole material which is in the 50,000-100,000 range. This giant-dielectric behaviour can be explained by an internal barrier layer (IBLC) at the grain boundaries. At last, the components of classical eddy current losses including losses due to ohmic effects and (true) dielectric losses on both bulk grain and boundary layers are distinguished.

I. INTRODUCTION

The constant reduction in size of switch mode power supplies leads to increase their working frequencies up to the MHz range. One of the main limitations in frequency increase is the energy dissipations by losses in ferrites that produce heating of the electronic circuits. So understanding and modelling the loss mechanism in Mn-Zn ferrite become more and more important. Recently, important works permits to theoretically separate the different components in losses, showing that eddy current losses become relevant in the MHz range[1,2]. Due to their complex mesostructure and related complex electrical behaviour, there are few data on the conductive properties of the sintered Mn-Zn ferrite, so it is difficult to model properly the eddy current phenomenon.
Eddy currents flowing into the conductive Mn-Zn ferrite produce losses by Joule effect. Nevertheless, the understanding of those conductive phenomenon is difficult because

polycrystalline ferrites are heterogeneous media where grains are connected to each other through boundary layers where some impurities are concentrated[3,4]. This leads to electrical properties drastically different compared to bulk grain. In fact, boundary region are known to increase both resitivity and permittivity of the material and they depend strongly on the frequency. Addition of insulating impurities[4] -such as CaO, SiO_2, Al_2O_3- which segregate at the grain boundaries, decreases the magnitude of macroscopic eddy currents leading to a strong reduction of losses.
It is known that the low resistivity of Mn-Zn ferrite bulk grains is due to simultaneous presence of ferrous (Fe^{2+}) and ferric (Fe^{3+}) ions on equivalent lattice sites which produces hopping electronic conduction[3]. Electrons jumping from ferrous to ferric ions are at the origin of electric conductivity and the phenomenon increases with larger Fe^{2+} contents. Their ohmic properties are assumed to be semi-conducting like, because around room temperatures, the conductivity increases with temperature. Furthermore, high dielectric properties exist in bulk ferrite grains with high relative permittivity[5].
In the other hand, the grain boundary resistivity is assumed to be several orders of magnitude higher than the bulk grain one. So, the mixed property leads to a material with enhanced resistivity but in the same time, the dielectric constant increases dramatically, sometimes as high as 50000 (in relative) or more. At last, Mn-Zn ferrites display a so-called “giant dielectric phenomenon” where the grain boundary (internal) barrier layers act like capacitances (IBLC) with very high values[6,7,8].
But, at high frequency, those IBLC short cuts the high resistance of the boundaries layers, so the global resistance of the ferrite decrease in a large part. The displacement currents[9] due to those shunting capacitors results in a strong increase of the eddy current losses in the MHz range. These losses are sometime referred as dielectric losses, but the term is misleading. Indeed, strictly speaking, dielectric losses are produced by electric dipoles relaxation in the lattice (linked charge current) which is different to ohmic losses (free charge current).
The aim of this work is to investigate the frequency dependent dielectric and conductive properties of the bulk grains and boundary layers separately. From an electrical point of view, the heterogeneous medium can be represented by an equivalent circuit. The bulk grains are equivalent to a resistance connected in parallel with a capacitance. The same model is used for the grain boundaries. At last, the two resistance-capacitance cells are connected in series and we obtain the equivalent electric scheme for the ferrite. AC complex resistivity of a commercial ferrite was measured and the parameters of the model were fitted. It appears that the resistance related to the bulk grain is 27 times lower than the ferrite global resistance at low frequency. Furthermore, the relative permittivity of the bulk grains is around 350 at low frequency which is strongly different from the boundary properties.
STEM (Scanning Transmission Electron Microscopy) and EDS (Energy Dispersion Spectrometry) revealed the calcium enrichment at the boundaries, that may explain the wide difference in electrical properties between bulk material and boundary layer.
At last, the obtained parameters of the ferrite properties are included into a simple model to distinguish the different sources of power dissipation when eddy currents flow.

II. EXPERIMENTAL PROCEDURE AND MEASUREMENT RESULTS

The Mn-Zn ferrite core under study is purchased from Ferroxcube (reference 3E27). It has been chosen for its low DC resistivity in order to enhance expected electric phenomenon. The resistivity measurement was conducted on a small bar shaped ferrite sample (l=6.4mm long and $S=1\times1mm^2$ of cross section) cut from an I rod (reference I/25/6/6-3E27 Ferroxcube). Skin effects become negligible in the sample, due to its small cross section area[10]. Furthermore, this geometry with large long/short axis ratio minimizes the value of the parasitic inductance which is proportional to µS/l. Secondly, given the ferrite electrical properties and due to the geometry of the cut, the impedance (in modulus) is in the range of 1500 Ω-50 Ω in the DC-500 MHz frequency band which permits high accuracy resistance measurement.

In order to ensure electrical contacts, some methods were tested such as silver conductive paste or silver conductive paint. But in all cases the electric contact between ferrite and conductive layers exhibit high resistances. Finally, 50 nm thick platinum electrodes with negligible contact resistances were deposited at each end of the 1×1 mm^2 bar using sputtering method.

The complex AC resistivity $\rho = \rho' + i\rho''$ was measured in the long direction of the sample using a HP4195A impedance analyser between 10 kHz and 500 MHz (circle in Fig. 1) at room temperature (~20°C). First, it appears that ρ' diminishes from 0.23 Ω.m at 10 kHz to 0.0085 Ω.m at 100 MHz. The latter value is assumed to be the intrinsic real part resistivity of the bulk grains because at this high frequency, the high resistance of the boundaries layers is short cut by its associated layer capacitance. So, it is seen that the boundary layers increase approximately 27 times the resistivity of the ferrite in the low frequency range. Secondly, the imaginary part of the resistivity, always negative, shows that the ferrite also behave as a capacitor as it was expected.

To estimate the influence of the temperature on the electric properties, DC resistivity measurement[11] were performed for temperature between 18°C and 33°C. The ferrite sample was placed in a temperature controlled chamber, and the DC resistance was measured using a 8 digits ohmmeter (Keithley 2700), in 4 wires configuration. The Mn-Zn ferrite behaves like an intrinsic semiconductor as its DC resistivity decreases with temperature around room temperatures (see in Fig. 2).

From the complex AC resistivity measurement $\rho^* = \rho' + i\rho''$ (where the * symbol denotes complex quantities), we have deduced the complex permittivity $\varepsilon^* = \varepsilon' - i\varepsilon''$ for the material supposed homogeneous as follows:

$$\varepsilon^* = \varepsilon' - i\varepsilon'' = \frac{\sigma''}{\omega} - i\frac{\sigma'}{\omega} \tag{1}$$

where $\sigma^* = \sigma' + i\sigma''$ is the complex conductivity obtained from the complex resistivity ρ^*, and ω is the angular frequency. The relative value of ε' at low frequency is around 8.3×10^4 which is consistent with data available in the literature[3]. This high value is due to the mixed properties of the heterogeneous structure of the sintered ferrite where the low resistivity bulk grains and the high capacitance of thin boundary layers (IBLC) act together. But these mixed dielectric properties could be much different from the intrinsic properties of each component taken separately.

III. THEORITICAL MODEL AND FITTING PROCEDURE

The complex impedance behaviour of the ferrite can be modelled by an equivalent circuit[12]. The bulk grains interior are equivalent to a resistance R_g (constant with frequency) connected in parallel with a capacitance C_g (see Fig. 3), where R_g is related to purely ohmic conduction phenomenon. The same model is used for the grain boundary layers where R_b and C_g are introduced. At last, the two resistance-capacitance cells are connected in series and we obtain the equivalent electric scheme for the ferrite. Here we assumed that the contacts between the ferrite and the two platinum electrodes are perfect, so it is not necessary to take them into account in the equivalent scheme. An inductance L is added in series to take into account inductive phenomenon that may happen at high frequency. In fact, due to the sample geometry (6.4 times longer than thick) and magnetic properties (relative permeability: ~4000 at low frequency) auto-induction phenomenon may modify the electrical impedance. But this effect is very low: $L\omega<10^{-5}\,\Omega$ at 0.5 GHz (see Table 1 for the L value) and can be disregarded.

From a general point of view, both grain core and boundary layers exhibit dielectric properties that may be due to various polarisation processes[13] (space charge, dipolar, ionic, atomic, electronic…). The contribution of each polarization mechanisms (and so the permittivity) depends on frequency. Ionic and electronic polarization processes are working for frequencies from DC up to 10^{12} Hz. For most oxide materials, their contribution to relative permittivity is in the 10 range. But it is known that bulk grain ferrite have anomalous high relative dielectric constant (in the 100 range) due to phenomenon not yet totally understand that cover ionic and electronic effects. Furthermore, an associated relaxation effect may occur in the MHz range that is due to time required for polarization (time lag between applied electric field and induced displacement current). Knowing the value of this time relaxation in the bulk grains, we will be able to discriminate among the different polarization mechanisms. So a complex dielectric permittivity $\varepsilon_g^* = \varepsilon_g' - i\varepsilon_g''$ is introduced to take into account dielectric relaxation, where real and imaginary parts strongly depend on the frequency. The dielectric dissipation factor or loss tangent factor $\tan\delta = \varepsilon_g'' / \varepsilon_g'$ expresses the ability of the dielectric material to convert electrical energy into thermal that produce heating of the material. But, from a physical point of view, these phenomena differ from the ohmic loss modelled by R_g and R_b (constant with the frequency) and we have chosen to model them separately. At last, to fit properly the impedance curves from 10 kHz to 500 MHz, it is necessary to consider the frequency dependences of permittivity and relaxation phenomenon for each material (bulk grains and boundaries). Furthermore, it is well known that impurities concentrated at the boundary leads to ohmic effect widely increase. Here, we want to investigate the effects of impurities on dielectric properties too, which can give some information on the anomalous polarization process that occurs in ferrite material up to the MHz range.
The most complete model for the complex permittivity including non-exponential relaxation phenomenon has been given by Havriliak and Negami[14] which is a combination of Cole-Cole and Cole-Davidson formula. For the boundary layer, it leads to:

$$\varepsilon_b^* = \varepsilon_b^\infty + \frac{\varepsilon_b^s - \varepsilon_b^\infty}{\left(1 + \left(i\dfrac{\omega}{\omega_b}\right)^a\right)^b} \tag{2}$$

where ε_b^* is the relative complex permittivity, ε_b^∞ is the optical dielectric constant related to the electronic polarisation process, ε_b^s is the static dielectric constant and ω_b is the angular relaxation frequency. Furthermore, exponents a and b are between 0 and 1, tacking into account the distribution of relaxation times.
The same law is used to model the dielectric behaviour of the bulk grain:

$$\varepsilon_g^* = \varepsilon_g^\infty + \frac{\varepsilon_g^s - \varepsilon_g^\infty}{\left(1 + \left(i\dfrac{\omega}{\omega_g}\right)^c\right)^d} \tag{3}$$

Here, it is assume that for both bulk grains and grain boundaries, the static dielectric constants are much greater than the optical ones. So equ. (2) and (3) can reduce to:

$$\varepsilon_b^* \approx \frac{\varepsilon_b^s}{\left(1+\left(i\frac{\omega}{\omega_b}\right)^a\right)^b} \tag{4}$$

$$\varepsilon_g^* \approx \frac{\varepsilon_g^s}{\left(1+\left(i\frac{\omega}{\omega_g}\right)^c\right)^d} \tag{5}$$

A ferrite sample with length *l* and square cross section of area *S* can be represented[4,12] as cubic grains with mean size D connected by boundary layers with mean thickness δ (see Fig. 4). In this case, the number n of grains in the long direction of the sample is:

$$n = \frac{l}{D+\delta} \approx \frac{l}{D} \tag{6}$$

because we assume that grains are much thicker than the boundary layers ($D >> \delta$). It follows that the total thickness of grains in the long direction is $nD \approx l$ with a good approximation. So the equivalent capacitance (now complex) for the bulk grains is given by:

$$C_g^* \approx \frac{S\varepsilon_g^s / l}{\left(1+\left(i\frac{\omega}{\omega_g}\right)^c\right)^d} \tag{7}$$

where *S* is the cross section area of the ferrite sample.

In general, it is very difficult to estimate the mean thickness δ of one boundary layer. So, here to avoid this problem, we express the equivalent capacitance for the grain boundary layers as a function of a static capacitance C_b^s:

$$C_b^* \approx \frac{C_b^s}{\left(1+\left(i\frac{\omega}{\omega_b}\right)^a\right)^b} \tag{8}$$

where $C_b^s = S\varepsilon_b^s / n\delta$ is proportional to the static permittivity.

Note that C_g^* and C_b^* are complex capacitance and their imaginary parts are related to the dielectric losses (relaxation type) in the two components of the ferrite.

At this step of calculation, we must clarify one point. Most authors in the literature introduce ohmic and dielectric losses in a global resistance connected in parallel with a capacitance having constant and real permittivity. This simple cell scheme is helpful to calculate by finite element method the eddy current flowing along complex paths in a ferrite core under AC magnetic flux up to the MHz range. In our approach, ohmic and dielectric losses are split to model properly the non-exponential dielectric relaxation phenomenon up to 500 MHz which

can give information on impurities influence on material properties. As a consequence, the equivalent capacitances C_g^* and C_b^* are complex and their real parts are related to dielectric loss and not Joule losses in the boundary layers.
The complex impedance Z^* for the electric equivalent scheme of a sample of length l and cross section area S is calculated as follow:

$$Z^* = iL\omega + \frac{R_g}{1 + iR_g C_g^* \omega} + \frac{R_b}{1 + iR_b C_b^* \omega} \tag{9}$$

The theoretical complex resistivity is then deduced from equ. (9):

$$\rho^* = \rho' + i\rho'' = Z^* S / l \tag{10}$$

This formula can be fitted to the complex resistivity data (circle symbols in Fig. 1) using the eleven fitting parameters ε_g^s, ω_g, c, d, R_g, C_b^s, ω_b, a, b, R_b, and L. Here we use a simplex algorithm[15,16] for finding the minimum value of a function. The squared difference between the data and the calculation with Eq.10 is the criterion to be minimized. Fig. 1 presents the measured complex resistivity spectra and the fitted theoretical curves given by Eq.(10) for frequency between 10 kHz and 500 MHz. It is seen that the measured data and the fitted theoretical curves match well and the fitted values are given Table I.
First, it appears that the equivalent resistance for the boundary layers, R_b, is 27 times much higher than the one for the bulk grains, R_g. This confirms the role of the grain boundaries in the DC resistivity behaviour of the ferrite. The bulk grain resistivity is around 0.0083 Ωm if we assume that D>>δ.
The estimation of a boundary layer DC resistivity is more difficult because the D/δ ratio must be known precisely according Eq.(11):

$$\rho_b^{DC} = \frac{R_b S}{n\delta} = \frac{R_b S}{l} \frac{D}{\delta} \tag{11}$$

where the number n of grain boundary layers along the thickness l is given by Eq. (6).
In the literature, there is few information on grains boundaries structure. Some papers[4,17] give a D/δ ratio in the range of 10^4. But this value strongly depends on chemical composition and sample preparation. So we have conducted SEM and STEM studies to understand the ferrite grains structure of our samples.

IV. SEM AND STEM STUDIES ON FERRITE STRUCTURES.

A grain diameter of D=17 μm in average has been deduced from scanning electron microscopy (Hitachi S-3400N SEM) analysis of the surface of the ferrite (see Fig. 5).
In order to investigate grain boundary structure and its chemical composition, STEM (Scanning Transmission Electron Microscopy) analysis was performed using a Tecnai F20 operating at 200 kV. Figure 6 shows an image of the ferrite obtained in bright field. The square indicates a small region (160x160nm^2) where two grains are separated by a boundary layer (dark region). EDS (Energy Dispersion Spectrometry) mapping was performed for this area in order to detect additives at the grains boundary. Figure 7 shows the EDS map for calcium detection. The change in contrast indicates that this element is concentrated in a region which corresponds to the boundary layer (bright region). Other possible additives (such as Na, K, Si) were not found in the boundary region (no contrast on the EDS map). An EDS line profile analysis were performed along the cross section of the boundary layer for the calcium (figure 8). This element is concentrated in a region of ~20 nm thick and it is assumed that this area match with the boundary layer from an electrical point of view because Ca addition dramatically increases the resistivity of the ferrite.

V. RESULTS AND DISCUSSION.

According to Table I (R_b=1359 Ω) and assuming δ~20 nm, it leads to a resistivity in the 10^{10} Ωm range for the boundary layer material which is very high compared to the bulk grain one (0.0083 Ωm). EDS studies shows that the chemistry of the two components are quite similar and they differ only in the Ca contents. It may argued that Ca^{2+} ions in the boundary layers take occupy some Fe^{2+} lattice positions, so electron jump of from ferrous to ferric ions is disrupted, so hopping phenomenon is quenched.

From the obtained fitted values (Table 1), and using Eq. (5) and (8), the dielectric properties (real part an imaginary part) of the bulk grains and the boundary layers are plotted for frequencies between DC and 500 MHz on Fig. 9 and 10. Note that extrapolation is done below 10 kHz. Havriliak and Negami model is a description of non-exponential relaxation based on the assumption that there is a distribution of relaxation times in subsystems. In Eq. (4) and (5), the constants *a* and *b* in one hand and *c* and *d* in the other hand are related to the shape of the relaxation times distribution in the boundaries and the bulk grains respectively. In spite of relaxation occurring in the same frequency range (MHz range), it appears that the real part of the permittivity of boundary layers decreases sharply for frequency over 100 kHz (Fig. 10), whereas for the bulk grain the real part of the permittivity decrease smoothly in the same frequency range (Fig. 9). It means that for the two materials the distribution of relaxation times are quite different, and it seems to be larger in the case of boundary layer material. The enlargement of relaxation time distribution might be due at least in part to the segregation of Ca impurities. Furthermore, bulk grains dielectric relaxations in the MHz range are not related to space charge polarisation effect (it works for frequency less than 10^4 Hz) or ionic and electronic polarisations (relaxation occurs at 10^{12} Hz). One can suppose that this anomalous polarisation mechanism may be related to hopping electronic phenomenon due to simultaneous presence of ferrous and ferric ions.

Compared to the DC relative permittivity of the sintered ferrite (8.3×10^4), dielectric properties of the bulk grains are very small ($\varepsilon_g^s = 354$ in relative). This conduct us to think that the boundary layers (IBLC) play an important role in the high apparent permittivity. The static equivalent capacitor is given Eq.(12) where the D/δ ratio is used again.

$$\boldsymbol{C}_b^s = \frac{\varepsilon_b^s \boldsymbol{S}}{\boldsymbol{n}\delta} = \frac{\varepsilon_b^s \boldsymbol{S}}{\boldsymbol{l}} \frac{\boldsymbol{D}}{\delta} \tag{12}$$

The dielectric constant is then express as:

$$\varepsilon_b^s = \boldsymbol{C}_b^s \frac{\boldsymbol{l}}{\boldsymbol{S}} \frac{\delta}{\boldsymbol{D}} \tag{13}$$

The D/δ ratio value is not exactly known, but taking it in the 10^3 range (using EDS and SEM measurement results: D~20 µm and δ~20 nm), one obtains the static relative permittivity for the boundary layers in the range of about 100. So, it should also be noted that the static dielectric constant of boundary layers are quite different from the bulk grains. In fact, Ca oxides contained in the powder are know to segregate under impurities forms in the boundary regions, producing at these places magnetic and dielectric properties far from that of ferrite crystallites[17,18,19]. Since the ferrite impedance is modelled by series resistance-capacitance elements, the anomalously high apparent dielectric constant of ferrites is a consequence of the high capacitance of the thin boundary regions connected in series by highly conductive bulk grains. Note that rather than the effect of the dielectric constant which is only about 100, the high capacitance of a boundary region is essentially due to its very low thickness (in the range of few tens nanometres).

Fig. 2 has shown that the sintered Mn-Zn ferrite behaves like an intrinsic semiconductor. Let us explain this phenomenon as a result of the mixed properties of bulk grains and boundary layers resistivities. The global DC resistance of the ferrite is given by:

$$\boldsymbol{R} = \boldsymbol{R}_g + \boldsymbol{R}_b \tag{14}$$

where $\boldsymbol{R}_g = 50\ \Omega$ and $\boldsymbol{R}_b = 1359\ \Omega$ (see Table 1) in the room temperature (T~20°C). Looking Fig. 2, one can calculate that the DC resistivity, and so the resistance *R*, decrease in a ratio of 12.5% if the temperature increase from 20°C to 30°C. It leads that the global resistance *R* decrease of about 176 Ω in this temperature range which is three times higher than the value of R_g at 20°C. One may conclude that the larger part of the semiconducting properties measured on the ferrite are due to the boundary layers (related to R_b). The semi conducting effects in the bulk grains, if they exist, are hidden because $R_g << R_b$. A thin layer of about 20 nm containing Ca impurities (quenching the hopping phenomenon between Fe^{2+} and Fe^{3+} ions) may behave like a semi conductor.

VI. EDDY CURRENT AND MAGNETIC LOSS CALCULATION.

Power losses in ferrite materials under ac excitation are associated with all irreversible phenomena taking place during core magnetization. Spin rotation damping, domain wall relaxation, and classical eddy current are the three main contributions of losses and at last, all of them are converted into heat. Eddy current field is dramatically non uniform in the cross section of the core and losses could be estimated only by electromagnetic field simulation. Here we have used finite element method (FEM) to compute losses in linear regime as it has been published in details in a recent paper[20]. The calculation is made for a bar shaped ferrite (6.4x6.4 mm^2 squared cross-section) where a sinusoidal excitation $\boldsymbol{H}_z(\boldsymbol{x},\boldsymbol{y},\boldsymbol{t}) = \boldsymbol{H}_0(\boldsymbol{x},\boldsymbol{y})\boldsymbol{e}^{\boldsymbol{i}\omega\boldsymbol{t}}$ is applied in the long direction (z axis). According to Maxwell equations, the diffusion equation of magnetic field was solved in two dimensions (in the xy plane) using a finite element code (under Matlab Pdetool):

$$\rho\nabla^2\boldsymbol{H}_0(\boldsymbol{x},\boldsymbol{y}) = \boldsymbol{i}\omega\mu\ \boldsymbol{H}_0(\boldsymbol{x},\boldsymbol{y}) \tag{15}$$

where $\rho = \rho' + \boldsymbol{i}\rho''$ is the complex resistivity of the ferrite material seen as homogeneous from an electrical point of view. It includes the mixed properties of bulk grains and grain boundary layers. On the other hand, $\mu = \mu' - \boldsymbol{i}\mu''$ is the complex permeability spectra measured (HP4195) on a toroidal ferrite sample with a section small enough to make classical eddy current and skin effect negligible. So, the μ'' parameter is related to local phase lag between **H** and **B** due to all dissipative phenomena (domain wall relaxation, spin damping…) except classical eddy current loss. At this step of calculation, we must clarify one point. In fact, ferrites have heterogeneous structure: high conducting grains are surrounded by near insulating boundary layers. Consequently, it exist local eddy currents constrained to circulate within the grains (excess eddy current loss). In other part, long-range paths eddy current flows from grains to grains through the boundary layers and at last it circulates within the macroscopic cross section of the magnetic circuit. This latter is usually named classical eddy current and it decreases with the reduction size of the magnetic circuit and become almost negligible (with respect to the other losses) if the magnetic circuit is small enough. Therefore, the local (excess) loss produced by eddy current constrained to circulate within the grains is included into the μ'' parameter but it is known to be negligible with respect to domain wall relaxation and spin damping losses.

The local magnetic loss field (including hysteresis magnetic loss and excess eddy current loss) related to the μ'' parameter can be written as:

$$p_{mag}(x,y)=\frac{1}{2}\omega\mu''\left|H_z(x,y)\right|^2=\frac{1}{2}\omega\frac{\mu''}{\left|\mu\right|^2}\left|B_z(x,y)\right|^2 \tag{16}$$

Taking the **H** field calculated with Equ.15 and from the fact that $j=\nabla\times H$, we can calculate the classical eddy current field across the core section. Note that j is the current density including the displacement and ohmic currents flowing from grains to grains through the boundary layers.

Knowing the current density field and considering Fig. 3, it is possible to estimate the different contributions to classical eddy current losses. It is obvious that ohmic losses exist for the bulk grains and boundary layers, related to the equivalent resistors R_g and R_b. The ohmic loss field in the bulk grains is computed using:

$$p_g^{ohmic}(x,y)=\frac{1}{2}R_g\frac{S}{l}\left|\frac{Z_{cg}}{R_g+Z_{cg}}\right|^2\left|j(x,y)\right|^2 \tag{17}$$

where $Z_{cg}=\frac{1}{iC_g^*\omega}$ is the impedance of the equivalent capacitance for the bulk grains, and $\left|j(x,y)\right|$ is the magnitude of the current density field.

In the same way, the power dissipated by ohmic losses in the boundary layers is:

$$p_b^{ohmic}(x,y)=\frac{1}{2}R_b\frac{S}{l}\left|\frac{Z_{cb}}{R_b+Z_{cb}}\right|^2\left|j(x,y)\right|^2 \tag{18}$$

where $Z_{cb}=\frac{1}{iC_b^*\omega}$ is the impedance of the equivalent capacitance for the boundary layers.

We must add dielectric losses to the ohmic ones. In fact, the current flowing in the equivalent capacitance C_g^* and C_b^* produce dielectric losses (truly speaking) due to the imaginary part of the dielectric constants related to the dipoles relaxation phenomena.

The power dissipated by dielectric losses in the bulk grains is:

$$p_g^{dielectric}(x,y)=\frac{1}{2}\mathrm{Re}\left(Z_{cg}\right)\frac{S}{l}\left|\frac{R_g}{R_g+Z_{cg}}\right|^2\left|j(x,y)\right|^2 \tag{19}$$

where Re refers to real part.

In the same way, the power dissipated by dielectric losses in the boundary layers is:

$$p_b^{dielectric}(x,y)=\frac{1}{2}\mathrm{Re}\left(Z_{cb}\right)\frac{S}{l}\left|\frac{R_b}{R_b+Z_{cb}}\right|^2\left|j(x,y)\right|^2 \tag{20}$$

At this point, we have calculated the different power loss fields which are inhomogeneous across the section. By averaging over the core section, we obtain the mean value of loss for all contribution:

$$<p>=\frac{1}{S_{ec}}\int_{-a/2}^{a/2}\int_{-a/2}^{a/2}p(x,y)dxdy \tag{21}$$

where S_{ec} is the square cross section area (S_{ec}=a×a) and a=6.4 mm.

At last, the loss per cycle per unit mass is obtained for each contribution:

$$w=\frac{<p>}{f\,\rho_m} \tag{22}$$

Where f is the frequency and ρ_m is the density of the ferrite material.

In Fig. 11, the different classical eddy current loss contributions are plotted as function of the frequency between 100 kHz and 3 MHz for a 50 mT peak induction. As expected, for frequency below 3 MHz, the power is mainly dissipated in the boundary layers by ohmic effect. Nevertheless, due to the corresponding high resistance R_b, and the small cross section area of the ferrite studied here (S_{ec}=6.4×6.4 mm^2), this power is low with respect to the magnetic loss. For frequencies over 400 kHz, dielectric loss in the boundary layers is the second contribution in eddy current loss. The contribution of the power dissipated by dielectric losses in the bulk grain is always negligible and it is not plotted in fig. 11.
Here, we have considered only the classical eddy currents, which are related to long-range patterns. Usually they are called dynamic eddy currents because they cancel for near static magnetic excitation. Nevertheless, it exist excess eddy currents surrounding the moving domain walls and at last they are confined within the grains. They are known to be negligible with respect to the classical (dynamic) eddy currents over the kHz region. But at very low frequency (quasi static condition), the Barkhausen jumps of domain walls produce excess (static) eddy currents that reach a nonzero value.
So, at very low frequencies, (excess) eddy currents are confined within the grains and energy dissipation is located in the grains by ohmic effect, whereas at frequencies over the kHz range, a global current circulation is established (classical eddy current) and dissipation is mainly located in the boundary layers by ohmic effect.
These calculations were compared in Fig. 12 to loss measurement obtained by a calorimetric method (described in details elsewhere[20]) on a 6.4×6.4×25.4 mm^3 bar shaped ferrite rod (3E27 material). The calculated eddy current loss is the sum of all bulk grains and boundary layers contributions:

$$w_{ec} = w_b^{ohmic} + w_b^{dielectric} + w_g^{ohmic} + w_g^{dielectric} \tag{23}$$

There is a good agreement between the measured loss (circle symbols) and the calculated total loss $w_{total} = w_{ec} + w_{mag}$ that include all possible local loss w_{mag} and loss w_{ec} due to classical eddy current that flows along macroscopic paths. It can be concluded that this computation method give good estimation in separation of each contribution (bulk grains and boundary layers) of classical eddy current loss.

VII. CONCLUSION.

In this study, we have presented a method to obtain the ohmic and dielectric properties of the two compounds constituting sintered Mn-Zn ferrite: the bulk grains and the boundary layers. The electric model used herein takes into account their ohmic and dielectric properties including relaxation phenomena. Theoretical formula was fitted on the impedance spectrum obtained on a ferrite sample with a good agreement for frequency between 10 kHz and 500 MHz. The deduced properties show that the two compounds have different ohmic and dielectric properties which are consistent with the STEM characterisation which shows a concentration of calcium at the boundary layers.
An important application of this characterisation method is the estimation of the different power dissipation sources when classical eddy currents flow in the bulk of a ferrite core. It appears that as expected for frequency less than 3 MHz, the classical eddy current losses are mainly dissipated in the boundary layers by ohmic effect. In an other part, the model reveal that for frequencies over 400 kHz, the dielectric losses in the boundary layers (due to the displacement current) have important influence in the level of eddy current loss.
In most cases, high resistivity Mn-Zn ferrites are obtained by adding some impurities such as silicon or calcium oxides, during the fabrication process. Ca, and Si which segregate at the boundaries increase the DC resistance. The present model can be helpful to quantify and

predict the effects of the additives on the complex resistivity which is at the origin of the power dissipated by eddy current. At last, the different contributions in eddy current power sources could be estimated.

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TABLE I. Values of fitted parameters.

Bulk grains parameters					Boundary layers parameters					Inductance
ε_g^s (relative)	f_g (Hz)	c	d	R_g (Ω)	C_b^s (F)	f_b (Hz)	a	b	R_b (Ω)	L (H)
354	2.82×10^6	0.386	0.892	50.2	1.31×10^{-10}	4.55×10^5	0.908	0.186	1359	1.42×10^{-14}

Figures Captions:

Fig.1 : real and imaginary part of complex resistivity. Circle symbols: measurement. Solid line: fitted curves obtained from the theoretical model.

Fig. 2: curve of DC resistivity versus temperature measured between 18°C and 33°C.

Fig.3: electric equivalent scheme for the sintered ferrite corresponding to the bulk grains (R_g, C_g) and the grain boundary layers grains (R_b, C_b).

Fig.4: structure of sintered ferrite including bulk grains of thickness D each, and grain boundary layer of thickness δ. Bulk grains mean diameter of around 20 μm have been deduced from a scanning electron microscopy analysis of the surface of the ferrite.

Fig. 5: Scanning electron microscopy analysis of the surface of the ferrite.

Fig. 6: STEM image showing the ferrite microstructure in a boundary region. The white line square corresponds to the area for the EDS maps.

Fig. 7: EDS mapping for the calcium. The bright area corresponds to a higher concentration of Ca.

Fig. 8: relative Xray counts in EDS analysis for Calcium detection along a cross sectional line.

Fig. 9: real and imaginary part of the bulk grain relative permittivity plotted as function of the frequency.

Fig. 10: real and imaginary part of the boundary layers equivalent capacitance plotted as function of the frequency.

Fig. 11: contributions in energy dissipated when a 50 mT alternative induction is applied in the ferrite. Solid line: energy dissipated by magnetic loss. Dashed line: dissipation by ohmic effect in the boundary layers due to eddy current. Dotted line: dissipation by dielectric loss in the boundary layers due to eddy current. Dashed-Dotted line: dissipation by ohmic effect in the bulk grains due to eddy current.

Fig.12: Energy loss for a 50 mT peak induction. Circles: measurement with a calorimetric method. Dashed line: calculated magnetic loss. Dotted line: calculated eddy current loss (w_{ec}). Solid line: calculated total loss (magnetic and eddy current losses w_{total}).