Mirowave Permeability of Ferrite Loaded Composites : Comparison Between Model and Experiment

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Abstract. Composite materials with various ferrite volume fractions were prepared by mixing powders of Ni-Zn ferrite in a dielectric matrix. Measurements of their electromagnetic properties were compared with the modeling results obtained with the aggregate theory. The comparison shows a good agreement between experimental and computed values.

1. INTRODUCTION

Compared with ceramic materials, composite materials present several advantages. They are easy to shape by molding and to machine. The computation of their electromagnetic properties from the knowledge of the properties of their constituents is still the subject of many studies. These properties depend on the permeability and the permittivity of the two phases, the volume fraction, the shape of the inclusions, the morphology of their spatial distribution, especially their state of aggregation. In the low frequency range (between one megahertz and one gigahertz) classical mixing laws do not accurately describe experimental results. In this paper, a new model based on the aggregate theory is proposed and applied to ferrite loaded composites with different volume fractions. The model and the experiments are restricted to the case of unpolarized materials: $H_{ext} = 0$.

2. PREPARATION AND CHARACTERISATION OF MATERIALS

The particulate phase was a nickel zinc ferrite powder $Ni_{0.3}\ Zn_{0.7}\ Fe_2\ O_4$ prepared by conventional ceramic processing. The raw materials (NiO, ZnO, Fe_2O_3) were first ball milled. After calcination at 1250°C the powder was sieved in order to obtain a grain size between 200 and 400 µm.The magnetic moment of the powder was measured and X-ray diffraction spectra were performed to control the phase formation. Single spinel phase was observed. Its magnetic moment was found equal to the one of the sintered material : 48 Gcm^3/g. SEM observation of the powder showed polycrystalline 200-400 µm spheres made of ceramic with an average grain size of 5 µm. This powder was then mixed with an epoxy resin and pressed into pellets with a pressure of 10^3 kg/cm^2. Several composites with volume fractions between 18% and 65% were prepared. After hardening of the resin, toroidal samples designed to be inserted into a standard 7 mm coaxial line were machined.
The measurements were performed between 100 MHz and 18 GHz. A network analyser was used. The complex permeability and permittivity were computed from the values of the measured reflection and transmission coefficients.

3. DESCRIPTION OF THE MODEL

We developed a new theory which is based in part on the work we have done for dielectric-conductor composites. In contrast to other theories, our approach takes fully into account the microscopic structure of the composite. Since fractal aggregates often occurs in such heterogeneous systems, we had to calculate their electromagnetic properties [1] then we used these results to calculate the electromagnetic properties of composites [2]. Since the problem is different for magnetic materials, we have to develop a new approach in this case.
First of all, we studied the permeability of the composites and of the bulk material by using Cole-Cole diagrams. This approach permitted us to understand the different contributions to the permeability. For bulk ferrites, one usually observes several maxima in the $\mu''(f)$ curve. These maxima can be ascribed either to wall domain vibration or spin rotation. By varying the microstructure and anisotropy field Ha in a series of materials, it is possible to separate these contributions [3]. In compo-

site materials, the situation is much less clear. Simple models based on mixing laws (such as Looyenga) enable to adjust some results [4], but they give no insight into the physics of these materials. The main problem comes from the fact that the resonance and in particular the ferrimagnetic resonance frequency in the composite are far from the ones in the bulk material.
The second and most important part of our work is to understand qualitatively and even quantitatively the difference between the resonance frequency of the composite and the bulk material. Our approach is based on the Polder and Smit model [5], which takes into account the effect of Weiss domain structure on the effective demagnetizing field in ferromagnetic resonance experiments on a multidomain particle. The particle is modelled as an ellipsoid divided into thin Weiss domains separated by 180° walls perpendicular to one of its principal axes. For a given particle with a regular arrangement of Weiss domains parallel to the plane xy, where Oz is taken as the local direction of the static magnetization Ms, with an effective anisotropy field Ha, the effective demagnetizing factors are different for a microwave field parallel to x (perpendicular to the Bloch walls) and a field parallel to y (parallel to the Bloch walls). Using Kittel's equation for an ellipsoid with demagnetizing factors α_x, α_y, α_z ($\alpha_x + \alpha_y + \alpha_z = 1$), one finds two extreme resonance frequencies :

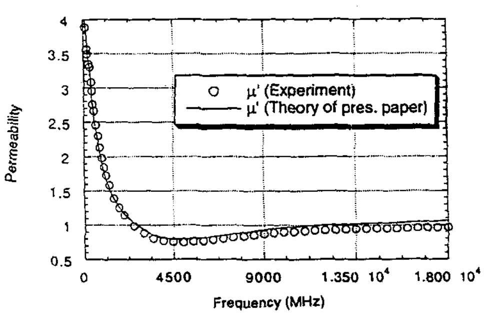
$$\omega_1 = \gamma\sqrt{H_a(H_a+4\pi M_s|\alpha_x-\alpha_y|)} \quad \text{and} \quad \omega_2 = \gamma\sqrt{(H_a+4\pi M_s)(H_a+4\pi M_s|\alpha_x-\alpha_y|)}\,.$$

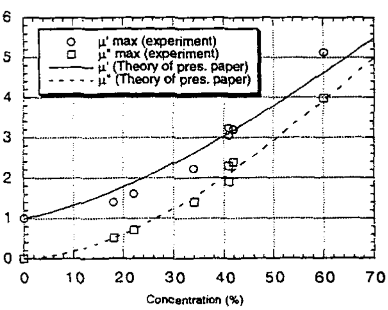
Since $H_a << M_s$, even for $(\alpha_x-\alpha_y) << 1$, H_a is still typically small compared to $4\pi\gamma M_s\sqrt{|\alpha_x-\alpha_y|}$.
The crucial point is the following. Both in the bulk material and in the spherical inclusions, there exist domains. Indeed the spheres are too large to remain a single domain. But the geometrical constraints on the spheres are much more stringent, thus the shapes of the domains have less freedom. Therefore the typical value of $|\alpha_x-\alpha_y|$ will be higher for the inclusions than for the bulk material, pushing ω_1 and ω_2 to higher values. In particular, Re(μ-1) remains positive up to much higher frequencies.
Finally, the resonance frequency in the composite material can be given by the expression :

$$\omega_{comp} \approx \gamma\sqrt{H_a 4\pi M_s|\alpha_x-\alpha_y|_{max}},$$ where $|\alpha_x-\alpha_y|_{max}$ is near unity, while for the bulk material $\omega_{bulk}=\gamma$ Ha as $|\alpha_x-\alpha_y|=0$.

This model permits us to compute the equivalent permeability μ_{inc} of the inclusion from the one of the bulk material.
For $4\pi M_s = 3200$ Oe, $H_a = 7$ Oe, we find a resonance for μ_{inc} at 420 MHz that is in agreement with the experiments.
The last step is to use the new "theory of aggregates" described in the beginning to calculate the properties of the composite using the permeability μ_{inc}.

4. RESULTS





The figures show a comparison of the permeability of the composite materials measured on samples and calculated by the present model. The left figure is a plot of the real part of the permeability versus frequency, for a given composite material with volume fraction 47%. The right figure shows the values of the complex permeability of a series of composite materials with different volume fractions measured for every material at its resonance frequency. Given the experimental error, estimated to be $\Delta\mu \approx 0.1$, these figures show an excellent agreement between theory and experiment.

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