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Experimental Study of Microwave Susceptibility in Ferrite Composite Materials

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Abstract. The microwave susceptibility of NiFe_2O_4 and Co_2Z was measured between 100 and 10000 MHz. The samples were either samples with different porosities or composites with different volume fractions, in the case of NiFe_2O_4 , or composites with different granulometry, in the case of Co_2Z . The measured susceptibility undergoes substantial changes at the crossover from a ceramic state to a composite of isolated particles and for different granulometries of Co_2Z in the range 10 to 600 μm . We show that a model in which the susceptibility is uniform inside the particles cannot account for these results. We suggest that in order to compute the susceptibility of composites, a microscopic approach is required.

1. INTRODUCTION

Ferrite composite materials have been the object of considerable interest for microwave applications^{1,2,3}. The polymer matrix, with its low permittivity, allows the microwave absorption band to be enlarged⁴. Another interest is for processing reasons, since it is easier to cover a complex shape with a compound polymer-ferrite than with a monolithic ceramic. In both cases, if we want to optimize the final material, we have to predict the permeability of composites or, at the very least, know the different parameters which can influence it. The behaviour at high frequency of the susceptibility is very different for composites and bulk materials. The aim of this paper is to stress the difficulties in modeling by two characteristic examples. In a first part, the influence of the volume fraction in NiFe_2O_4 composites is presented and discussed. In a second part, we compare these results with the influence of granulometry in Co_2Z composites.

2. CONCENTRATION DEPENDENCE

NiFe_2O_4 was prepared by sintering reaction. Four samples were obtained in one heat treatment (1250°C for 24 hours) with different porosities depending on the uniaxial pressure applied on the oxide powders: one bulk sample with less than 4 % porosity, two porous samples with 28 % and 39 % porosity and one powder sample. The latter was lightly milled to be sieved under 25 μm and then dispersed in a PMMA (polymethylmetacrylate) matrix with 55% and 28 % volume fraction. The susceptibility of bulk NiFe_2O_4 , porous NiFe_2O_4 and composites was measured between 100 and 10000 MHz using a HP8510 network analyser on toroidal samples placed in a coaxial wave guide at the APC7 standard.

The behaviour of the susceptibility (χ') is given on figure 1. Two features stand out. First, for slightly the same volume fraction (composite with 55 % volume fraction and porous ceramic with 62 % volume fraction), the level of magnetic losses generated at low frequency ($f < 1000$ MHz) is much lower in the composite than in the ceramic. Secondly, the natural spin resonance is strongly shifted to higher frequencies, from 1800 MHz for bulk NiFe_2O_4 to 4500 MHz for the composite with 28 % volume fraction.

In trying to explain these results, we have used an effective medium approach. The susceptibility of an isolated particle χ_{ext} , made of a material with intrinsic susceptibility χ_i , embedded in a medium of susceptibility χ_m is given by the relationship:

$$\chi_{ext} = \frac{\chi_i}{1 + N(\text{shape}, \chi_i, \chi_m) \cdot \chi_i} \quad (1).$$

N , the demagnetizing factor, is a function of particle shape, χ_i and χ_m . In the effective medium approach, the influence of other magnetic particles is taken into account by taking χ_m as the susceptibility of the composite χ . Since χ is a function of the volume fraction f , the demagnetizing factor N is also a function of f . With $\chi = f \cdot \chi_{ext}$, the susceptibility of the composite is given by the relationship:

$$\chi = \frac{f \cdot \chi_i}{1 + N(\text{shape}, \chi_i, f) \chi_i} \quad (2).$$

Neglecting variations of particle shape throughout a real sample and magnetostatic interactions between particles, formula (2) is a test of a model in which the magnetic domain configuration is ignored.

Inverting relation (2), we compute N for heterogenous materials, given the volume fraction, the susceptibility χ of composites and the susceptibility χ_i of the bulk ferrite. The result, shown on figure 2, displays two regimes, with a crossover between 1000 and 2000 MHz.

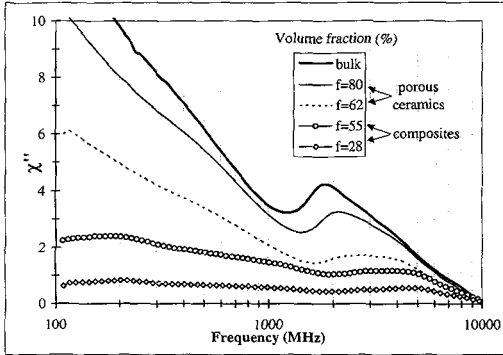


Figure 1- Imaginary part of the susceptibility as a function of volume fraction.

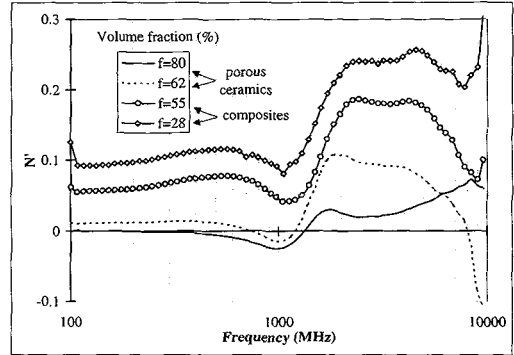


Figure 2- Real part of N , obtained from (2). The imaginary part of N is always small except between 1 and 2 GHz.

In both regimes, N' is roughly constant with frequency and increases when the volume fraction decreases. However, N' takes two distinct values in the two regimes. This cannot be explained by the dependence of N with χ_i (the frequency dependences of N and χ_i is very different). Thus, the two values reflect two different microscopic magnetization mechanisms: as is known, below 1000 MHz, the susceptibility comes from domain wall motion while above 2000 MHz, it comes from the natural spin resonance. To explain the difference between bulk samples and composites, the microscopic character of these mechanisms must be described.

3. GRANULOMETRY DEPENDENCE

The influence of granulometry was also studied for the planar hexaferrite Co_2Z ($\text{Ba}_3\text{Co}_2\text{Fe}_{24}\text{O}_{41}$) prepared by coprecipitation⁵. Three granulometries were obtained ($\phi < 25 \mu\text{m}$, $50 < \phi < 100 \mu\text{m}$, $200 < \phi < 600 \mu\text{m}$) by sieving and the powders were dispersed in a polymer matrix to obtain samples with the same volume fraction of 30 %.

The measured susceptibility on these samples is presented on figure 3. It shows a strong dependence with granulometry which was not observed for NiFe_2O_4 in the same granulometry range. This phenomenon cannot be accounted by the approach of the previous paragraph, the demagnetizing factor N being independent of particle size. The influence of granulometry indicates a different configuration of magnetization inside particles of different size.

4. CONCLUSION

We have shown that for, NiFe_2O_4 , the relation between the intrinsic susceptibility (bulk sample) and the susceptibility of composites depends on the dissipation mechanism (domain wall motion or gyromagnetic). For Co_2Z , the dependence of susceptibility with granulometry cannot be explained by a simple model in which magnetic structure is ignored. In both cases, in order to explain the experimental results, the topography of magnetization at a lengthscale below the particle size has to be described. To this end, we are now developing micromagnetic computations⁶.

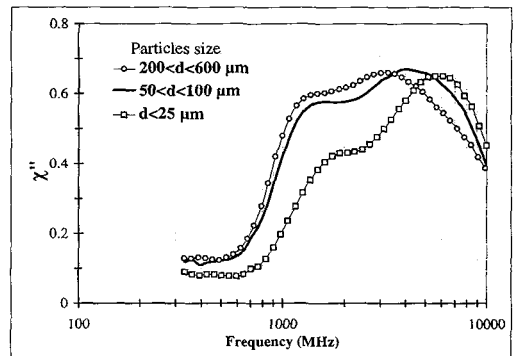


Figure 3: granulometry dependence of the susceptibility in composites Co_2Z -polymer.

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