STUDY ON THE DIELECTRIC PROPERTIES OF THE FERROFLUIDS IN HIGH FREQUENCY FIELD

N. Stefu[a], P. C. Fannin[b], I. Malaescu[a], C. N. Marin[a]

[a] *West University of Timisoara, Faculty of Physics, Bd. V. Parvan, no. 4, Timisoara, Romania*

[b] *Department of Electronic and Electrical Engineering, Trinity College, Dublin 2, Ireland.*

Abstract
Measurements of the real and the imaginary components of the complex dielectric permitivity for a ferrofluid consisting of magnetite particles surfacted with oleic acid, dispersed in kerosene, and for the ferrofluid constituents, in the frequency range of 100MHz – 6GHz are presented
The paper presents existing theoretical models regarding the dielectric properties of a binary dielectric mixture and, based on the experimental measurements, verifies the Maxwell – Garnet and the Gunther-Heinrich models for a ferrofluid considered to be a dielectric binary system.

1. Introduction

Ferrofluids, or magnetic liquids, are colloidal suspension of ferro- or ferrimagnetic nanoparticles (diameter of aproximately 10 nm) dispersed in a liquid matrix, stabilized with a compatible surfactant (organic most of the times). The ferrofluids are inhomogeneous media, consisting of three elements: the basic liquid, the magnetic particles and the stabilizer. The dielectric properties of the ferrofluids depend on several factors such as: chemical composition, obtaining method, volume ratio, particles dimensions, temperature, presence of a magnetic field, etc [1, 2, 3]. The present paper studies the dielectric properties of ferrofluids in high frequency fields, considering the ferrofluids dielectric heterogeneous media, based on the Maxwell – Garnet and the Gunther-Heinrich models.

1.1 *Dependence of the permitivity on the frequency*

The dielectric permitivity of the ferrofluid is a complex, frequency dependent quantity, which can be written, $\varepsilon(\omega) = \varepsilon'(\omega) - j\varepsilon''(\omega)$, where ε'(ω) represents the real part, while ε"(ω) represents the imaginary part of the complex permittivity.

Besides the Debye model [4], there are some other equations for the frequency dependencies of the complex permittivity, such as the Cole-Cole dependence [5] (eq.1), and the Havriliak - Negami dependence [6] (eq. 2):

$$\varepsilon(\omega) = \varepsilon'(\omega) - j\varepsilon''(\omega) = \varepsilon_\infty + \frac{\varepsilon(0) - \varepsilon_\infty}{1 + (j\omega\tau)^{1-\alpha}} \qquad 0 \le \alpha < 1 \tag{1}$$

$$\varepsilon(\omega) = \varepsilon'(\omega) - j\varepsilon''(\omega) = \varepsilon_\infty + \frac{\varepsilon(0) - \varepsilon_\infty}{\left[1 + (j\omega\tau)^{1-\alpha}\right]^\beta} \qquad \begin{array}{l} 0 \le \alpha < 1 \\[6pt] 0 < \beta \le 1 \end{array} \tag{2}$$

In these equations, ε_∞ is the high frequency permitivity value, $\varepsilon(0)$ is the static permitivity (or at very low frequencies), $\omega = 2\pi f$ is the angular frequency, and τ is the relaxation time.

1.2. The effective permitivity of heterogeneous dielectrics

A heterogenous medium represents a space region arbitrary filled with a mixture of various dielectric materials. The computation of the dielectric permitivity of a medium consisting of a mixture of different dielectric materials is an issue of great practical and theoretical interest. But the determination of the response of such a medium to the action of an electric field is a very difficult problem, even if information on the dielectric behavior of the constituents and their spatial distribution is available. The dielectric behavior of a dielectric mixture can be modeled using an effective dielectric permitivity, ε_{eff}, which represents a spatial weighted average of the permitivities of the constituents [7]. Because of the difficulty of the problem, binary mixtures (mixtures with two dielectric components) are mostly studied.

The most known equation for the relative effective permitivity of a binary dielectric mixture is the Maxwell - Wagner equation [7, 8], obtained for systems consisting of dielectric spheres (ε_1) dispersed in a dielectric medium (ε_2). Another method, proposed by Rayleigh, and then generalized by Gunther and Heinrich [7, 9] to describe the permitivity of a sistem consisting of spheres (ε_1) randomly dispersed in a continuous dielectric medium (ε_2), gives the equation:

$$\varepsilon_{eff} = \varepsilon_2 \cdot \frac{\left[(1-K) - 2v\dfrac{\varepsilon_2 - \varepsilon_1}{2\varepsilon_2 + \varepsilon_1}\right]}{\left[(1-K) + v\dfrac{\varepsilon_2 - \varepsilon_1}{2\varepsilon_2 + \varepsilon_1}\right]} \tag{3}$$

where K is a correction factor depending on the precise way the spheres are dispersed in the medium (the maximum value is K=0.25), and v is the volume ratio, representing the ratio of the total volume of the spheres on the volume of the system.

Another usual equation is the Maxwell-Garnet equation [10]:

$$\varepsilon_{eff} = \varepsilon_2 + 3v\varepsilon_2 \frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + 2\varepsilon_2 - v(\varepsilon_1 - \varepsilon_2)} \qquad (4)$$

These equations make no reference to the frequency of the electric field they only refer to the static permitivity of the composite. On the other hand, the above equations do not take into account the surface interactions between the particles and the dispersion medium [11].

The present paper investigates whether these equations fit to the experimental values of the dielectric permitivity of the ferrofluid (mixture of nanodimensional spheres, surfacted, dispersed in a dielectric liquid) taking into account the frequency dependency of the dielectric permitivity of the components. A term giving the electric conductivity of the ferrofluid was added to equations (3) and (4), resulting in the equations

$$\varepsilon_{eff} = \varepsilon_2 \cdot \frac{\left[(1-K) - 2v\frac{\varepsilon_2 - \varepsilon_1}{2\varepsilon_2 + \varepsilon_1}\right]}{\left[(1-K) + v\frac{\varepsilon_2 - \varepsilon_1}{2\varepsilon_2 + \varepsilon_1}\right]} - j\frac{\sigma}{\omega \cdot \varepsilon_0} \qquad (5)$$

and

$$\varepsilon_{eff} = \varepsilon_2 + 3v\varepsilon_2 \frac{\varepsilon_1 - \varepsilon_2}{\varepsilon_1 + 2\varepsilon_2 - v(\varepsilon_1 - \varepsilon_2)} - j\frac{\sigma}{\omega \cdot \varepsilon_0} \qquad (6)$$

2. Measuring method

The sample studied was a ferrofluid consisting of magnetite particles, surfactated with oleic acid, dispersed in kerosene. The particles were obtained using the chemical coprecipitation methode, by hydrophobization in the absence of the dispersion medium [12]. The volume ratio was $v_A=0.13$. The static measurements were performed using the ollowing galvanometer methode, and the ollowing values were obtained: the saturation magnetization $M_\infty= 49.154 \ kA/m$ and the mean magnetic diameter $d_m= 8.5 \ nm$.

The dielectric properties of the ferrofluid were measured using the Hewlett-Packard analyzer and the transmission coaxial line [13]. The investigated frequency range was (100 MHz – 6 GHz). The dielectric properties of the materials that are the ferrofluid constituents have also been investigated: (i) the magnetite particles surfacted with oleic acid and (ii)

kerosene, using the same experimental technique. All measurements were performed at room temperature.

3. Results and discussion

3.1 The experimental data for the complex permitivity of the ferrofluid constituents

The ferrofluid samples can be considered binary dielectric mixtures having the surfacted particles and the kerosene as constituents. The experimental values for the permitivity of the surfacted magnetite particles are presented in figure 1 and the experimental values for the permitivity of the kerosene are presented in figure 2.
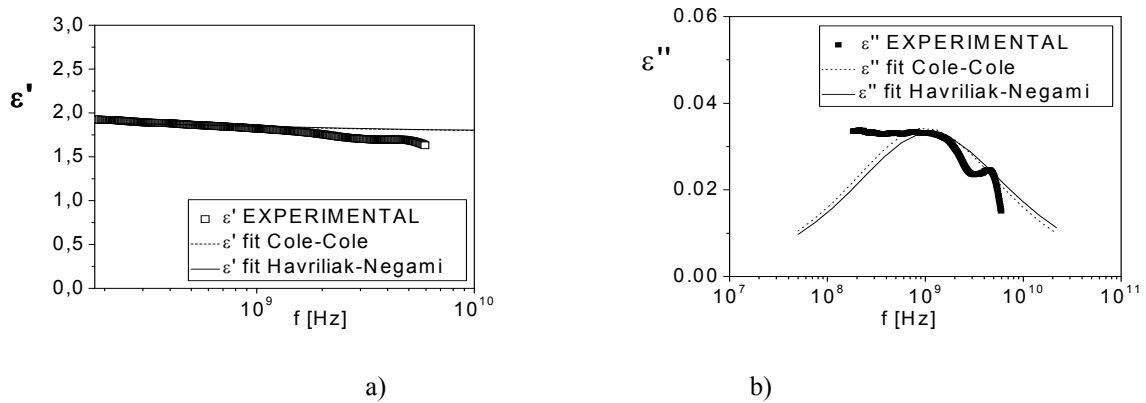

a)   b)

Fig. 1. Plot of the real component (a) and the imaginary component (b), of the relative permitivity of surfacted magnetite particles against f(Hz)
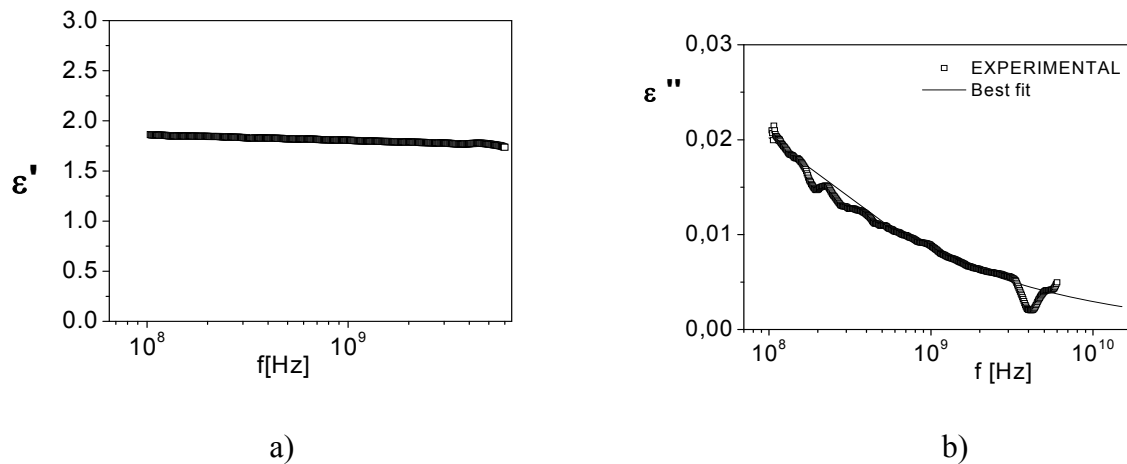

a)   b)

Fig. 2. Plot of the real component (experimental values) (a) and the imaginary component (b), of the complex permitivity of kerosene against f(Hz).

The experimental data for the surfacted magnetite particles in figure 1, have been fitted using Cole-Cole (1) and Havriliak − Negami (2) equations, and the corresponding fitting parameters α_{CC} (Cole − Cole), α_{HN} and β_{HN} (Havriliak-Negami) were obtained.

$$\alpha_{CC} = 0.29; \quad \alpha_{HN} = 0.29 \text{ si } \beta_{HN} = 0.92 \qquad\qquad (7)$$

It can be seen in figure 2 that the real component ε' of the complex permitivity of kerosene is almost independent of frequency, having a constant value of 1.8, while the imaginary component ε'' decreases with the frequency.

3.2 The complex effective permitivity of the ferrofluid. Theoretical data and comparison to the experimental data

We have computed the frequency dependence of the complex effective permitivity of a ferrofluid, considering it as a heterogeneous dielectric medium, using the equations (5) and (6), obtained from the Gunther-Heinrich (GH) (3), and Maxwell – Garnet (MG) (4) equations respectively, by adding a conductivity term. In these equations we considered the quantities ε_1 and ε_2 as complex quantities and frequency dependant, using the experimental values:

- ε_1 represents the complex permitivity of the magnetite surfacted particles, (figure 1)

- ε_2 represents the complex permitivity for kerosene (figure 2).

The equations (5) and (6) were used to compute the theoretical values for the effective permittivity of the ferrofluid, which are presented in figure 3, along with the experimental data for the studied ferrofluid sample for comparison.

For the ferrofluid conductivity the value $\sigma = 0.2 \cdot 10^{-6}$ $(\Omega \cdot m)^{-1}$ [14] was used, and the parameter $K = 0.25$ was considered, as indicated in [15] for spherical particles.

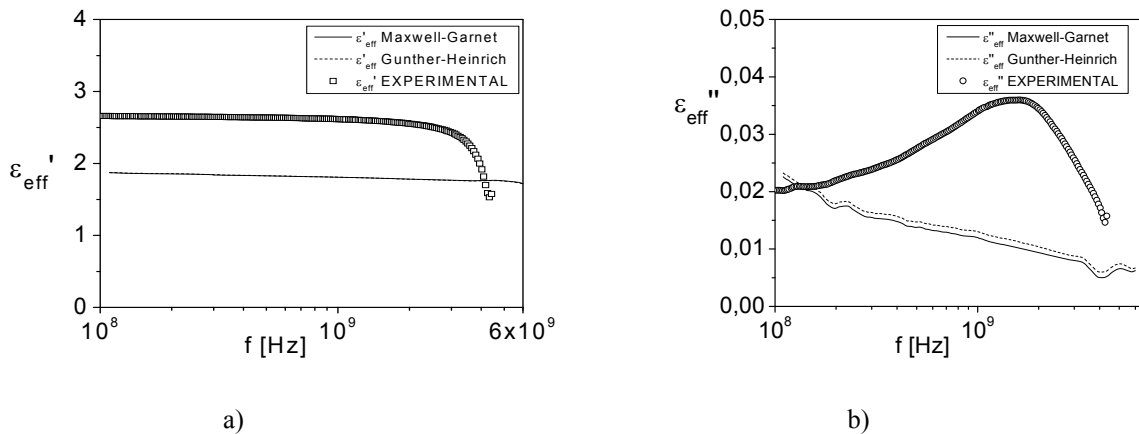a)   b)

Fig. 3. The effective permitivity for the ferrofluid sample, computed and experimental data

From figure 3 a) it can be noticed that all used models give approximately the same values for ε'_{eff}, the computed values being lower than the experimental ones. From figure 3 b) it can be seen that there are some differences between the experimental data and the theoretical values for ε''_{eff}, for both equations studied. These differences show that both

Maxwell-Garnet and Gunther-Heinrich models do not fit to the experimental data obtained for a ferrofluid in the investigated frequency range, if the ferrofluid is considered to be just a binary dielectric mixture having the surfacted particles and the kerosene as constituents. The differences that occur between the computed and the experimental data are probably due to the fact that the effective medium equations do not take into account the interfacial phenomena and the dipole-dipole interactions, and, on the other hand, traces of electrolyte may have remained inside the ferrofluid due to the obtaining method and ions may be adsorbed on the surfactant layer on the particles.

Further research will be conducted in order to explain the observed differences and to propose new equations to model the ferrofluid dielectric behaviour in high frequency field.

4. Conclusions

The investigated sample was a magnetic fluid with magnetite particles surfacted with oleic acid, dispersed in kerosene. Measurements of the real and the imaginary components of the complex dielectric permitivity for the sample and for the ferrofluid constituents, in the frequency range of 100MHz – 6GHz are presented

Assuming the ferrofluid a binary dielectric mixture of two components (the surfacted magnetite particles and the kerosene), we compared the computed values given by the theoretical Gunther-Heinrich and Maxwell – Garnet equations to the experimental data obtained for the ferrofluid. Some differences between the experimental data and the theoretical values for the effective permitivity, for both theoretical equations are observed.

We may assume that these differences are due to the interfacial phenomena and the dipole-dipole interactions, and to the traces of electrolyte that may have remained inside the ferrofluid.

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