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Measurement of Complex Permittivity of Oil-Based Ferrofluid in Magnetic Field

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The changes of dielectric parameters in oil-based ferrofluid have been measured in an external magnetic field. The frequency dependent real permittivity and the dissipation factor were measured within the frequency ranges from 1 mHz to 2 MHz by a capacitance method. These parameters have been studied in combined electric and magnetic field, when fields were parallel and perpendicular. The Cole–Cole model has been used to analyze measured data. When a magnetic field was applied, the interaction between the magnetic field and magnetic moments of nanoparticles led to the aggregation of magnetic nanoparticles to new structures — thick chains which had influence on the value of dielectric permittivity. At constant magnetic field the dependence of real permittivity and $\tan\delta$ on angle between the electric and magnetic field (anisotropy) were measured, too. The various influences of magnetic field development on the investigated liquid are discussed.

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1. Introduction

Ferrofluids have specific properties and various technical and biomedical fields of their applications already exist. The study of their dielectric parameters by dielectric spectroscopy can lead to better understand polarization phenomena and improve their properties. The measurements for various concentrations and temperatures showed two main relaxation processes [1, 2], the first for high frequencies — the Maxwell–Wagner effect and the second for low frequencies — the Schwarz model [3]. The dielectric behavior of ferrofluid changes with the application of an external magnetic field and with the relative orientation of the electric and magnetic fields. This effect is known as magneto-dielectric anisotropy effect [2, 4]. The acoustic spectroscopy is also very useful tool for the study of structure changes of ferrofluids in the magnetic fields [5, 6].

The complex relative permittivity is one of important dielectric parameter, which is a measure of how much energy from an external electric field is stored in a material and how dissipative a material is to an external field. It can be modeled by the Cole–Cole model plus the addition of dc losses [7] in the form

$$\varepsilon^* = \varepsilon_\infty + \frac{\varepsilon_s - \varepsilon_\infty}{1 + (\mathrm{j}\omega\tau)^\alpha} + \mathrm{j}\frac{\sigma_{dc}}{\omega\varepsilon_0}, \tag{1}$$

where σ_{dc} is dc conductivity, α is empirical exponents, τ is time constant of relaxation polarization, ε_s is static dielectric constant, ε_∞ is optical constant and ω is angular frequency of the electric field.

2. Experimental

The studied substance was the ferrofluid EMG-909 with particle diameter (d = 10 nm) dispersed in light hydrocarbon oil, the volume concentration of magnetic particle in ferrofluid was 3.8%. The ferrofluid parameters are: viscosity 3 mPa s, density 1.02 g/cm^3, initial magnetic susceptibility 1.38 and magnetic saturation 22 mT. In our experiment we have used liquid crystal (LC) cells as capacitors to achieve more precise dielectric measurement of ferrofluids. In the LC cells, two indium tin oxide (ITO) conductive, transparent, thin layers function as electrodes. Liquid crystal cells with two parallel plate ITO electrodes, whose distance apart $d = 100$ μm and the active electrode area was $A = 60$ mm^2, were used as capacitors (sample holders). The capacitance of the air filled cell was $C_0 = 2.6$ pF. The whole system was placed on the thermally stabilized block by water from thermostat JULABO F25. In order to investigate the influence of a static magnetic field on the electro-kinetics in the ferrofluid, the capacitor was placed in electromagnet.

The dielectric parameters: real capacitance and resistivity by LCR Meter OT 7600 Plus at frequencies from 100 Hz to 2 MHz and real and imaginary capacitances by IDAX 350 from 1 mHz to 10 kHz were measured. All measurements were done at temperature 25 °C.

3. Results and discussion

The real relative permittivity of the ferrofluid measured in the absence and presence of the magnetic field is depicted in Fig. 1. The real permittivity increases with decrease of frequency of the electric field. Its values also increase with temperature. The complex permittivity measured by IDAX is higher than 20 for frequencies below 1 Hz so these values are not shown in Fig. 1. In the measured frequency range, the real permittivity spectrum depended on the orientation of magnetic and electric field. The permittivity of the ferrofluid is increased for parallel and decreased for perpendicular orientation only for frequencies lower than 4 kHz. The variations of the dielectric permittivity with the applied magnetic

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field are a result of rearrangement of nanoparticles into new structures — chains aligned in direction of magnetic field [6]. The induced anisotropy in real permittivity of a ferrofluid subjected to an external magnetic field can be seen.

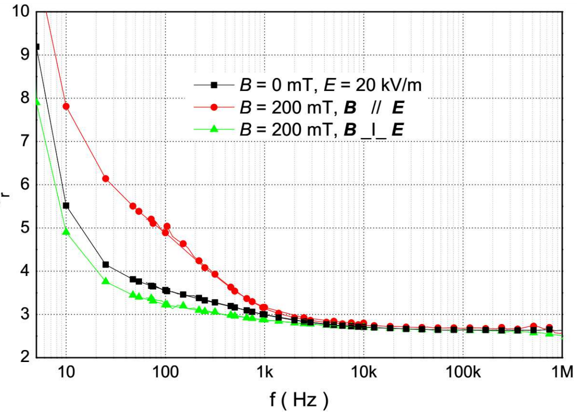

Fig. 1. The dependence of the real permittivity on the frequency on various magnetic field and its orientation to the electric field.

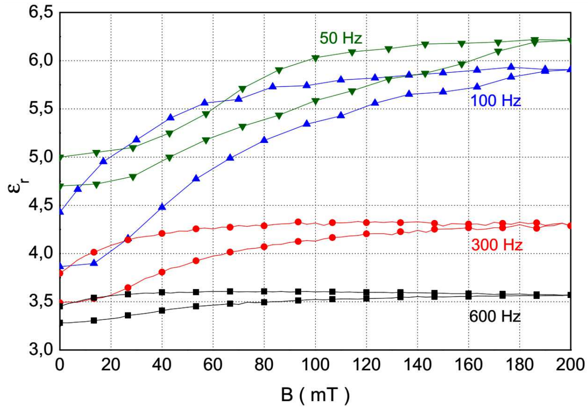

Fig. 2. The dependence of the real permittivity in linear change of external magnetic field to 200 mT with time step 3.3 mT/min for various frequency of the electric field. Electric and magnetic field are parallel.

In the low frequency range (below 1 kHz), we can see the pronounced dielectric dispersion, which is related to a relaxation process [1]. Since the ferrofluids belong to the complex systems, a continuous distribution of relaxation times can be expected. The analysis of low frequency relaxation process was done by fitting the complex permittivity data with the Cole–Cole model [2, 7, 8]. The parameters in Eq. (1) were estimated by means of the least squares techniques to obtain the best fit for dielectric responses of the ferrofluid and they are listed in Table I. DC conductivity of ferrofluid slightly increased in magnetic field. This could be a result of new structures created in magnetic field, which have higher charge and general they increase the conductivity. Static dielectric constant is not in Table I, because its value depends on the selection of minimal input frequency at fitting by the Cole–Cole model. For the low frequency relaxation process, the best fitting value for α was close to 1, which gives the well-known Debye relaxation law. From calculated parameters we consider that the relaxation maximum is associated with a single relaxation process which can stem from a polarization of electric double layer observed in the magnetic particles [9] The Schwarz model of electric double layer polarization can be used to explain the low frequency relaxation maximum.

TABLE I

Parameters of ferrofluid in various magnetic fields estimated by Cole–Cole model at the electric field $E = 20$ kV/m.

Mag. field	0 mT	$\parallel$ 200 mT $\perp$	
σ_{dc} [nS/m]	54	59	64
ε_∞ [F/m]	2.49	2.99	2.44
τ [s^{-1}]	2.21	2.65	2.27
α	0.95	0.96	0.96

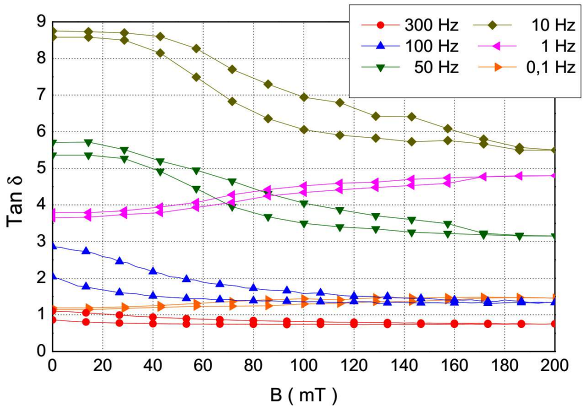

Fig. 3. The dependence of the dissipation factor in linear change of external magnetic field to 200 mT with time step 3.3 mT/min for various frequency of electric field. Electric and magnetic field are parallel.

The dependence of the real permittivity and the dissipation factor of ferrofluid on the magnetic field show a hysteresis effect (Figs. 2, 3). The same effect was measured also by acoustic spectroscopy [6]. The magnetic field is linearly increased from 0 mT to 200 mT for 1 h and decreases again with the same rate. The visible changes of the dielectric parameters with the magnetic field were observed at lower frequencies. The relative permittivity slightly increases with the increase of magnetic field. This effect is caused by the process of particle agglomeration and chains formation, which reduces the distance between the electrodes and the capacity (respectively real permittivity) growth. With the decrease of magnetic field, the process of structural changes can even continue and the real permittivity is almost constant until the magnetic field is high enough (600 Hz — 40 mT, 300 Hz — 80 mT, 100 Hz — 140 mT, 50 Hz — 160 mT). Then the real permittivity further decreases at the decrease of magnetic field, but it does not return to its initial value at zero field. This effect can be described by existence of chains, whose lifetime was longer than time of decrease of the magnetic field. The change of dissipation factor with linear change of the magnetic field is presented in wider spectrum of frequencies (Fig. 3). The dissipation factor decreases with the increase of magnetic field for frequencies higher than 10 Hz and at lower frequencies it increases. This effect is caused with local maximum of $\tan\delta$ dependent on frequency of the electric field and on the orientation and value of magnetic field ($B = 0$ mT: 5 Hz; $B = 200$ mT: 3.5 Hz for $\boldsymbol{E}\|\boldsymbol{B}$, 7 Hz for $\boldsymbol{E}\perp\boldsymbol{B}$.

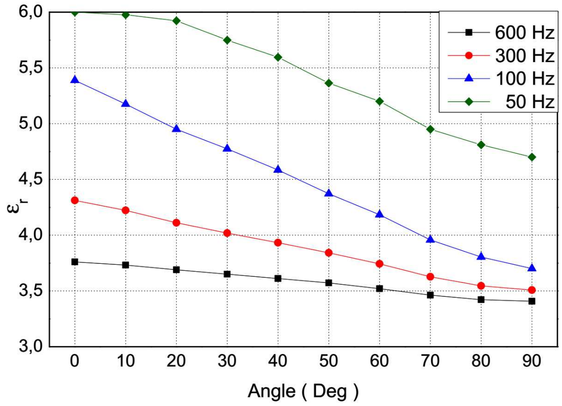

Fig. 4. Anisotropy of the real permittivity measured at $B = 200$ mT for various frequencies.

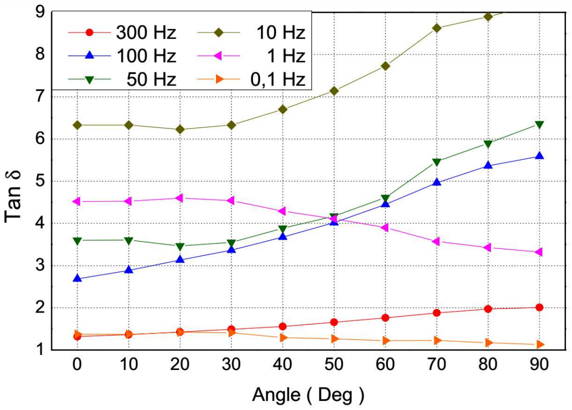

Fig. 5. Anisotropy of the dissipation factor measured at $B = 200$ mT for various frequencies.

Figure 4 shows the dependence of the anisotropy or magneto-dielectric effect of the real permittivity on the angle between the vectors of the electric and magnetic fields [2, 4]. The fields are parallel for angle 0° ($\boldsymbol{E}\|\boldsymbol{B}$). We observe that the real permittivity monotonically decreases with increase of angle. From this measurement we could suppose that only short chains are created in the direction of magnetic field. These simple structures have no additional effect on monotonic decrease of real permittivity because they have only translational motion in the direction of magnetic field. If these structures would be bigger — clusters, their rotational motion must be taken in account and the monotonic decrease of ε_r with angle could be changed, a local maximum will appear at angle around 45° [6]. For perpendicular orientation of the electric field to magnetic field Lorentz force has maximum value. This force causes nanoparticles motion on a spiral, which in final increases tracks of nanoparticles in the system and it is connected with decrease of permittivity. The anisotropy is also observed for dissipation factor (Fig. 5). Its maximum move to higher frequencies, so there is increase of its values for frequencies higher than 10 Hz with the increase of angle and for lower frequencies we observe decrease.

4. Conclusions

Dielectric properties of the ferrofluid in the applied magnetic field have been studied. Particle agglomeration within ferrofluid can be distinguished from dielectric spectroscopy and the magnetodielectric effect was observed. It has been shown that values of the complex permittivity and the dissipation factor change with the linear increase of the magnetic field. From the anisotropy measurement and the monotonic decrease of real permittivity with angle resulted that thick chains of nanoparticles are created in ferrofluid.

Acknowledgments

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