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Influence of Temperature on the Magneto-Dielectrics Effect of Oil-Based Ferrofluid

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The effect of temperature on the magneto-dielectrics behavior was studied by dielectric spectroscopy in the transformer oil-based ferrofluid with magnetic nanoparticles upon the effect of an external magnetic field. The frequency dependence of complex permittivity and dissipation factor were measured within the frequency range from 1 mHz to 10 kHz at different temperatures by a capacitance method. The dielectrics parameters were measured as a function of the external magnetic field in the range of 0–200 mT, parallel to the direction of the electric field as a function of temperature in the range of 15–35 °C. The interaction between magnetic field and magnetic moments of nanoparticles led to the aggregation of magnetic nanoparticles to new structures which had influence on dielectric parameters. The dependence of these parameters at constant magnetic field on angle between the direction of the electric and magnetic fields (anisotropy) has been measured, too.

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1. Introduction

Ferrofluids with specific properties have wide applications in various technical and biomedical fields. The study of their parameters and behavior under electric and magnetic fields can be done by various methods. The acoustic spectroscopy is one of useful tool for the study of structure changes of ferrofluids in the magnetic fields [1, 2]. However, nanoparticles are polarizable the dielectric spectroscopy can be used to study of their dielectric parameters. From measurement of these parameters can be describe the mechanisms of dielectric relaxations in ferrofluids. These dielectric relaxations can be seen as local maxima's on the frequency dependence of dissipation factor. One maximum appears at hundreds of MHz frequency, which exhibit in many colloidal systems containing different dielectric constituents is describe on the basis of Maxwell–Wagner model. The next low frequency relaxation process is the result of the creation of the electric double layers (EDLs) due to the presence the space charge in the ferrofluid, the Schwarz model [3–5]. EDLs are created on the particle surfaces as a result of electrostatic interactions between the particles and the space charge. Finally, the electrode polarization (EP) causes an anomalous increase in the complex dielectric permittivity in a lower frequency region by many orders of magnitude [4].

The dielectric behavior of ferrofluid depends also on the application external magnetic field and on the relative orientation of the electric and magnetic fields. This effect is known as magneto-dielectric anisotropy effect [6].

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2. Experimental

The studied substance was the ferrofluid with magnetic (Fe_3O_4) nanoparticle diameter 7.9 ± 2.4 nm dispersed in transformer oil (MOGUL TRAFO CZ-A, Paramo) with the volume concentration of magnetic particle 0.45%. The ferrofluid parameters are: viscosity 14.8 mPa.s, density 0.865 g/cm^3, DC magnetic susceptibility 0.06 and magnetic saturation 2.32 A m^{2}kg^{-1}.

The dielectric parameters of studied ferrofluid were measured by the capacitance method, where usually liquid crystal (LC) cells are used as capacitors (sample holders). In these cells Indium Tin Oxide (ITO) conductive, transparent, thin layers function as electrodes. The distance of two parallel plate ITO electrodes was $d = 5$ μm and the active electrode area was $A = 30$ mm^2. The capacitance of the air filled cell was $C_0 = 56$ pF. The whole system was thermally stabilized by thermostat JULABO F25. The dielectric parameters of ferrofluid were measured by IDAX 350 in the frequency range from 10 mHz to 10 kHz. In order to investigate the influence of a magnetic field on the dielectric parameters of the ferrofluid, the capacitor was placed in electromagnet. All measurements were done at three temperature 15, 25 and 35 °C.

3. Results and discussions

The development of $\tan\delta$ as a function of the frequency at various temperatures is depicted in Fig. 1. The main important parameter is the local maximum at an eigenfrequency $f_e(\omega_e)$, which is associated with the relaxation process of polarization. The Schwarz model [3, 5] of electric double-layer polarization can be used to explain this maximum in this range of frequencies. For $f/f_e \gg 1$ and $f/f_e \ll 1$, $\tan\delta$ tends to be equal to zero. With increasing temperature $\tan\delta$ and eigenfrequency f_e shift towards higher frequencies (15 °C – 30 Hz, 25 °C – 47 Hz, 35 °C – 68 Hz), due to the polarization of dielectric. The effect of the electrode polarization for this type of ferrofluid was not observed.

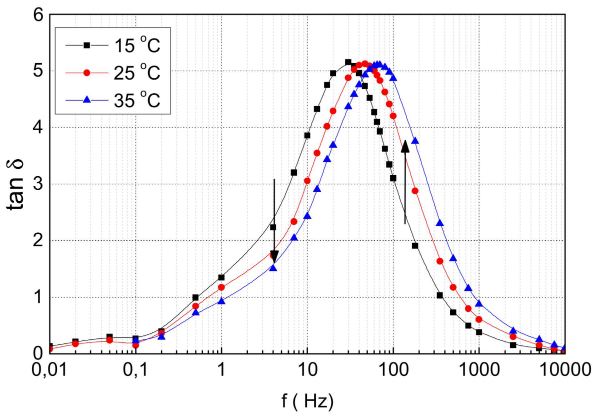

Fig. 1. Dependence of the dissipation factor on the frequency of the electric field at various temperatures. The full lines represent the fits of experimental data at each temperature.

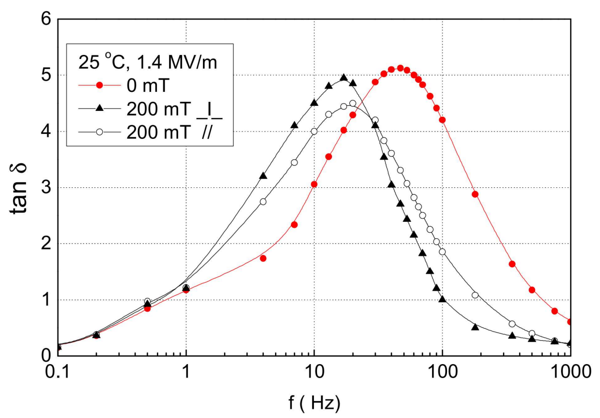

Fig. 2. Dependence of the dissipation factor on the frequency without and at the magnetic field 200 mT for two different orientations to the electric field measured at 25 °C.

The magneto-dielectric effect, the role of the magnetic field on the frequency dependence of dissipation factor of ferrofluid is presented in Fig. 2. The dissipation factor decreases under application of magnetic field. The magnetic field changes the structure of ferrofluid by rearranging the magnetic nanoparticles to forms like oligomers, thin or thick chains [7–9]. For the parallel orientation of the magnetic and electric fields the decrease is bigger than for a perpendicular orientation. The frequency dependence of dissipation factor in parallel orientated fields is only a function of the electric field, since the magnetic field has a minimal effect on the drift of the nanoparticles in the direction of the electric field. For the perpendicular orientation of the electric field to the magnetic field, the Lorentz force has a maximum value. This force causes the motion of nanoparticles in a spiral, which causes longer tracks of the nanoparticles in the system and is connected with the increase at the dissipation factor. The position of the local maximum of tan δ is also dependent on the orientation of the magnetic field with respect to the electric field and the temperature. For the magnetic field 200 mT, with a parallel orientation to the electric field, the maximum is at the frequency of 20 Hz. For a perpendicular orientation of the fields, the local maximum of tan δ occurs at little lower frequency 17 Hz.

The development of dissipation factor at two frequencies (lower frequency 7 Hz and higher one 100 Hz as the eigenfrequency) with increased magnetic field is shown in Fig. 3. The main changes are observed just after application of magnetic field, practically only to 80 mT. There is big increase at 7 Hz or decrease at 100 Hz with the increasing magnetic field. This effect is caused by a shift in the position of the local maximum of tan δ with the increase of the magnetic field. For magnetic fields higher than 80 mT almost no change are visible. At decrease of the magnetic field tan δ is almost constant. The dielectric losses in the dielectric increases with increasing temperature and the local maxima of tan δ shifts to higher frequencies (Fig. 1). This fact causes the increase of dissipation factor at higher frequency [9] and its increase at lower frequency with temperature.

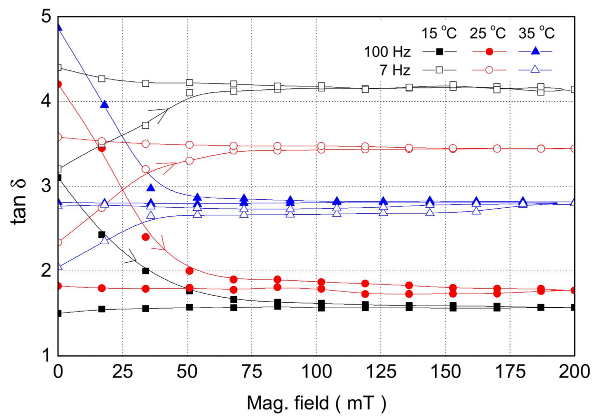

Fig. 3. Temperature dependence of the dissipation factor on the linear increase and decrease of the external magnetic field measured with a time step of 3.3 mT/min.

The analysis of the development of tan δ at given frequency and temperature depends on whether the frequency is lower or higher than the eigenfrequency. The influence of the magnetic field on the dielectric properties at frequencies lower than f_e is the opposite of that for higher frequencies. At higher frequencies, the electric dipole moments of created structures are unable to follow the change in the electric field intensity, and thus the effect of polarization has a smaller effect and the dissipation factor decreases with a linear increase of the magnetic field. At lower frequencies, the effect of polarization must be taken into account and it increases with the magnetic field. The effect that $\tan\delta$ does not return to its initial value at return to zero magnetic field can be explained by the existence of structures, which have a lifetime which is longer than the time of the decrease of the magnetic field.

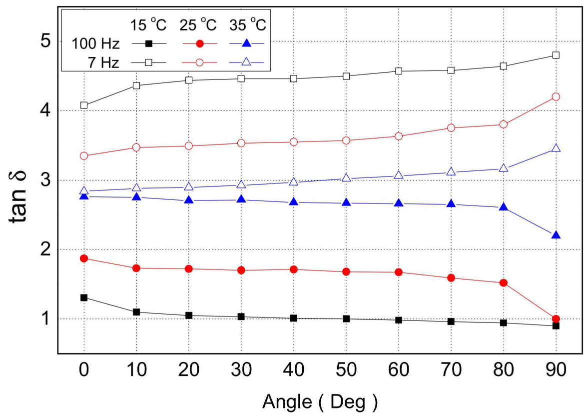

Fig. 4. The anisotropy of dissipation factor measured in the constant magnetic field 200 mT at frequencies 100 Hz and 7 Hz for three different temperatures.

On the basis of obtained results investigating the magneto-dielectric effect presented in Fig. 2, the anisotropy measurement was done. At this measurement, the magnetic field first jumps from 0 to 200 mT and then after the stabilization time, the angle between the electric and magnetic fields was changed. It can be seen (Fig. 4) that $\tan\delta$ at frequency 100 Hz decreases monotonically with increasing angle and at lower frequency 7 Hz there is an opposite effect. However, temperature also can affect the anisotropy measurement. The increase (100 Hz) or decrease (7 Hz) of their values were observed, although the character of development does not change.

The induced anisotropy in dissipation factor of the ferrofluid subjected to the external magnetic field can be seen. Figure 4 shows the dependence of the anisotropy or the magneto-dielectric effect of dissipation factor on the angle between the vectors of electric and magnetic fields. We observe that the dissipation factor changes monotonically with increasing angle [10]. From this measurement, it can be assumed that only short chains are created in the direction of the magnetic field. These simple structures have only translational motion in the direction of the magnetic field so they have no additional effect on a monotonic change in the dissipation factor. If these structures were bigger, i.e. clusters, their rotational motion would have to be taken into account and a local maximum will appear at anisotropy measurement.

4. Conclusions

Dielectric properties of the ferrofluid in the applied magnetic field were studied. Particle agglomeration within the ferrofluid was identified using dielectric spectroscopy and the magneto-dielectric effect was observed. It is shown here that the values of dissipation factor change with a linear change of the magnetic field. The results of the anisotropy measurement and the monotonic change of dissipation factor with angle show that only chains of nanoparticles are created in the ferrofluid.

Acknowledgments

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