DYNAMICAL PROPERTIES OF TRANSFORMER OIL BASED MAGNETIC FLUIDS

RAJNAK¹ M., TOTHOVA² J., KOVAC¹ J., KURIMSKY² J., DOLNIK² B., KOPČANSKÝ¹ P., MOLCAN¹ M., ROYER³ F., TIMKO¹ M. ¹Institute of Experimental Physics, Slovak Academy of Sciences, Watsonova 47, 040 01 Kosice, Slovakia, <u>timko@saske.sk</u> ²Faculty of Electrical Engineering and Informatics, Technical University, Kosice, Slovakia ³Laboratoire LT2C 25, rue du Dr Remy Annino 42000 Saint Etienne, France

Abstract: In this article, our experimental study of the dynamic dielectric behavior of transformer oil-based ferrofluid with magnetite nanoparticles is presented. Electro-kinetic properties of the original transformer oil MOGUL based ferrofluid sample with the magnetic volume fraction of 6.6 % were investigated by means of dielectric spectroscopy method. Frequency-dependent dielectric permittivity was measured within the frequency range from 20 Hz to 2MHz by a capacitance method. The effect of temperature on the viscosity of ferrofluid was studied using a modular compact viscosimeter. The temperature dependence of the viscosity was measured in the temperature range from 20 up to 80°C. The magnetization of different concentrations of MPs in MFs was determined by using of the vibrating sample magnetometer.

1. Introduction

Ferrofluids are colloidal suspensions of nanosized ferromagnetic particles coated with surfactants and dispersed in a carrier liquid. Since their physical properties can be easily influenced by external forces such as the magnetic field, they have found many applications in a variety of fields such as electronic packing, mechanical engineering, aerospace or bioengineering. In particular, if MFs are based on water as a carrier liquid, they can be used in such areas as magnetic drug delivery, cancer treatment by means of magnetic induced hyperthermia, or magnetic imagine [1]. One of many unique properties of ferrofluids is their tuneable viscosity by the external magnetic field. This is called the magnetoviscous effect [2]. One of the most successful applications of thermophysical property of magnetic fluid is audio speaker. In the audio speakers, magnetic fluid is filled around the voice coil. Because thermal conductivity of magnetic fluid is much larger than that of the air, the fluid provides a lower heat resistance between the coil and pole plate [3].

However, if we apply a magnetic fluid to heat transfer applications such as cooling system for power transformer or micromachine [4], it is necessary to perform the detailed investigation on the properties of heat transfer of a magnetic fluid under magnetic field. For a good understanding of this phenomenon, the knowledge of the viscous properties of MFs in the absence of magnetic fields is very important. Particularly its dependence on the amount of suspended magnetic particles (MPs) and temperature is very interesting.

Several previous studies [9-10, 14] on polarization and relaxation processes in ferrofluids were carried out using a dielectric spectroscopy method. It has been shown that the variation of both relative permittivity and dissipation factor with frequency may result from: (1) polarization due to molecular rotation either in polar liquids or in solid polar liquid mixtures, (2) polarization due to charge accumulation at the interfaces of different media in colloidal suspension, (3) polarization due to ion atmosphere displacement, and (4) polarization due to diffusion coupling between ion flows [15]. This indicates that the frequency dependence of relative permittivity and dissipation factor of ferrofluids permits identification and analysis of a number of completely different underlying mechanisms.

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. With the distances between the electrodes down to a micrometer range, only a low voltage is needed to obtain a high electric field and just a droplet of ferrofluid is required to fill the LC cell. The objective of this work is to examine magnetic, viscosity and dielectric properties of transformer oil-based ferrofluid placed in a LC cell under the electric field within the frequency range from 20 Hz to 2MHz.

1. Results

The measurement of magnetization was performed at room temperature by a vibrating sample magnetometer (VSM) with the uncertainty of about 1%. The saturation magnetization of the prepared undiluted ferrofluid was 26.6 A.m²·kg⁻¹. As it was shown earlier the shear stress versus shear rate dependence for pure transformer oil MOGUL is linear what is presenting a Newtonian-like fluid behavior. Similarly prepared sample of magnetic fluid based on this oil and used in our experiment behave as Newtonian fluids, since its viscosity do not depend on the shear rate too (results are not given here). The temperature dependence of viscosity shows the classical behaviour at which increasing temperature initiates decreasing viscosity for our sample of ferrofluid. The influence on viscosity is coming from changing the viscosity of pure oil with temperature and from Brownian motion of nanoparticles. With the increasing temperature the Brownian motion of the particles in the ferrofluid was strengthened, which reduced the speed difference between the carrier liquid and the magnetic particles.

Electro-kinetic properties of the original ferrofluid sample with the magnetic volume fraction of 6.6 % were investigated by means of dielectric spectroscopy method. To obtain the frequency dependent complex permittivity of the sample we employed an LCR meter (Agilent E4980A) with the frequency range from 20 Hz up to 2 MHz. Liquid crystal cells with two parallel plate ITO electrodes, whose distance apart $d = 1.6 \mu m$ and the active electrode area $A = 25 \text{ mm}^2$, were used as capacitors (sample holders). The capacitance of the air filled cell C_0 was 148 pF. In order to investigate the influence of a static magnetic field on the electro-kinetics in the ferrofluid, the capacitor was placed between two permanent squared magnets, separated 5 cm apart. The sample was therefore exposed to the quasi homogenous magnetic field of 150 mT. In that way the capacitance $C(\omega)$ and dissipation factor tan $\delta(\omega)$ of the sample were measured at room temperature. The related real and imaginary part of the complex permittivity was determined according to (1) and (2), respectively. The uncertainty of the acquired data is less than 0.3 %.

$$s'(\omega) = \frac{c(\omega)}{c_0}$$
(1)

$$s''(\omega) = s'(\omega) \tan \delta(\omega)$$
(2)

The complex permittivity of the undiluted ferrofluid measured in the absence of magnetic field is depicted in Fig. 1. In the low frequency range, one can see the pronounced dielectric dispersion, which is related to a relaxation process [5]. Since the ferrofluids belong to the complex systems, a continuous distribution of relaxation times can be expected. To analyze the low frequency relaxation process, we start by fitting the complex permittivity data with Havriliak-Negami equation [6, 7]:

$$s^*(\omega) = s_{\infty} + \frac{e_s - e_{\infty}}{[1 + (i\omega\tau_m)^{\alpha}]^{\beta}}$$
(3)

where α and β are empirical exponents, τ_{mi} is the characteristic relaxation time, ω is the angular frequency of the electric field, ε_{s} and ε_{∞} are the low and high frequency limits of the permittivity, respectively. For the low frequency relaxation process, the best fitting value of

the both empirical exponents was found to be 1, what gives the famous Debye relaxation law. Following these fitting parameters we consider the relaxation maximum to be associated with a single relaxation process which can stem from a polarization of electric double layer (EDL) presented on the magnetic particles. The possible formation of the EDL, consisting of adsorbed OH⁻ and oleate ions on the particle surface and surrounded by hydrated NH⁺ ions, is discussed in recent studies [8-11]. Such a relaxation is then described by Schwarz model of EDL polarization [12].



Figure 1: Frequency dependent complex dielectric permittivity.

In the measured frequency range, the real permittivity spectrum has been found as nearly independent on the magnetic field applied to the sample in parallel and perpendicular configurations in regard to the electric field (Fig. 2).



Figure 2: Influence of magnetic field on the real permittivity spectrum.



Figure 3: Magnetic field dependent imaginary permittivity spectrum.



Figure 4: The influence of magnetic field on the spectrum of dissipation factor.

The well-known magneto-dielectric effect [13-14] has not been directly proven in the ferrofluid micro layer. However, looking at the loss spectra (Fig. 3, 4), one can see the decrease in both, the magnitude of the loss peak and the relaxation time, when the magnetic field is applied. As the height of the relaxation maximum is associated with the value of static permittivity, one can deduce the decrease in the static permittivity as well. This is related to reduction in the electric dipole moment values and its ordering with E field. Then, the reduced distance between the ions forming the electric dipoles and their reduced relaxation path, when the magnetic field induces formation of chain like particle clusters, can account for the observed lower relaxation maxima. Nevertheless, better experimental statistics and more quantitative analysis is necessary to understand the non-typical magneto-dielectric phenomenon.

3. Conclusion

We have shown, that the temperature dependence of viscosity shows the classical behaviour at which increasing temperature initiates decreasing viscosity coming from changing the viscosity of pure oil with temperature and from Brownian motion of nanoparticles. With the increasing temperature the Brownian motion of the particles in the ferrofluid was strengthened, which reduced the speed difference between the carrier liquid and the magnetic particles. It was shown that the height of the relaxation maximum is associated with the value of static permittivity what can be assigned to the decreasing in the static permittivity. This fact is connected with reduction in the electric dipole moment values and its ordering with E field and formation of chain like particle cluster in E field too.

Acknowledgements.

This work was supported by the project VEGA 0043, 2/0045/13, 1/0487/12, the Slovak Research and Development Agency under the contract Nos. APVV-0171-10 and 20-006005, Ministry of Education Agency for Structural Funds of EU in frame of projects 26110230061, 26220120046 and M-era.Net – MACOSYS.

4. References

[1] Q.A. Pankhurst, N.K.T. Thanh, S.K. Jones, J. Dobson, J. Phys. D: Appl. Phys. **42**, 224001 (2009). DOI:10.1088/0022-3727/42/22/224001

[2] S. Odenbach, Magnetoviscous Effects in Ferrofluids ISBN 3-540-43068-7, Springer-Verlag, Berlin, 2002.

[3] D.B. Hathaway, Sound Eng. Mag., 13 42 (1979).

[4] K. Nakatsuka, B. Jeyadevan, S. Neveu, H. Koganezawa, J. Magn. Mag. Mat., 252, 360 (2002)

[5] Y. Feldman, A. Puzenko, and Y. Ryabov, Dielectric Relaxation Phenomena in Complex Materials, Fractals, Diffusion, and Relaxation in Disordered Complex Systems, W. T. Coffey and Y. P. Kalmykov, Eds. John Wiley & Sons, Inc., 2005, pp. 1–125.

[6] N. Axelrod, E. Axelrod, A. Gutina, A. Puzenko, P. B. Ishai, and Y. Feldman, Dielectric spectroscopy data treatment: I. Frequency domain, Meas. Sci. Technol., vol. 15, no. 4, p. 755, Apr. 2004.

[7] M. Wübbenhorst and J. van Turnhout, Analysis of complex dielectric spectra. I. One-dimensional derivative techniques and three-dimensional modelling, J. Non-Cryst. Solids, vol. 305, no. 1–3, pp. 40–49, Jul. 2002.

[8] I. Malaescu and C. N. Marin, Dielectric behavior of some ferrofluids in low-frequency fields, J. Colloid Interface Sci., vol. 251, no. 1, pp. 73–77, Jul. 2002.

[9] M. M. Rădulescu, Low-frequency dielectric losses in ferrofluids containing magnetite particles in kerosene, J. Magn. Magn. Mater., vol. 85, no. 1–3, pp. 144–146, Apr. 1990.

[10] P. C. Fannin, C. N. Marin, I. Malaescu, and N. Stefu, Microwave dielectric properties of magnetite colloidal particles in magnetic fluids, J. Phys. Condens. Matter, vol. 19, no. 3, p. 036104, Jan. 2007.

[11]M. Rajnak, J. Kurimsky, B. Dolnik, K. Marton, L. Tomco, A. Taculescu, L. Vekas, J. Kovac, I. Vavra, J. Tothova, P. Kopcansky, and M. Timko, Dielectric response of transformer oil based ferrofluid in low frequency range, J. Appl. Phys., vol. 114, no. 3, p. 034313, Jul. 2013.

[12] G. Schwarz, "A theory of the low-frequency dielectric dispersion of colloidal particles in electrolyte solution1,2" J. Phys. Chem., vol. 66, no. 12, pp. 2636–2642, Dec. 1962.

[13] A. Espurz, J. M. Alameda, and A. Espurz-Nieto, Magnetically induced dielectric anisotropy in concentrated ferrofluids, J. Phys. Appl. Phys., vol. 22, no. 8, p. 1174, Aug. 1989.

[14] A. Spanoudaki and R. Pelster, Frequency dependence of dielectric anisotropy in ferrofluids, J. Magn. Magn. Mater., vol. 252, pp. 71–73, Nov. 2002.

[15] K. Arulanandan, Low frequency dielectric dispersion of clay-water-electrolyte systems, Clays Clay Miner. 16, 337–351 (1968).