

Jozef Kudelcik - Peter Bury - Peter Kopcansky - Milan Timko *

TEMPERATURE EFFECT ON ANISOTROPY OF ACOUSTIC ATTENUATION IN MAGNETIC FLUIDS BASED ON TRANSFORMER OIL

Magnetic nanoparticles in magnetic fluids under the effect of magnetic field create structures, which are not arranged uniformly. These structures have various shapes dependent on the intensity and duration of applied magnetic field. In the case of such structures the anisotropy of acoustic attenuation can be observed. The effect of temperature on this process was studied by acoustic spectroscopy in transformer oil-based magnetic fluids. Taketomi theory that assumes existence of spherical clusters was used. These clusters form long chains, aligned in magnetic field direction. The cluster radius, the number density of the colloidal nanoparticles as well as other parameters of magnetic nanoparticles were determined from experimental results. These data allowed to draw conclusions about the changes in the parameters describing the structures of magnetic nanoparticles in the investigated magnetic fluid caused by different temperatures.

Keywords: Magnetic fluid, transformer oil MOGUL, anisotropy of acoustic attenuation.

1. Introduction

Nanotechnology is very progressive research area and their results can be found in many parts of our daily life. Nanoparticles are used in chemistry, biology, technology and other areas to improve the properties of various materials. One of the technological applications is utilization of magnetic nanoparticles in the magnetic fluids based on transformer oil [1 and 2]. These types of the transformer oils should have better insulating and thermal properties. The dielectric breakdown strength of these liquids is influenced by the concentration of magnetic nanoparticles and can have the positive influence on electric breakdown [3, 4 and 5].

One of the methods to study changes in the magnetic fluid structure is based on the measurement of the acoustic wave attenuation changes $\Delta\alpha$ under the external magnetic field at various conditions [6, 7 and 8]. The investigation of the attenuation changes of the acoustic wave propagating through suspensions, in which magnetic nanoparticles representing one phase are dispersed in carried liquid representing a continuous second phase, can indicate characteristic properties and structure of magnetic liquids. The interaction between the acoustic waves and the magnetic nanoparticles or clusters leads to the additional attenuation of acoustic wave compared to that in the carried liquid.

Several authors studied the arrangement of magnetic nanoparticles in magnetic fluid under the effect of an external magnetic field both theoretically and experimentally [2, 8 and 9]. There are also computer simulations [10 and 11] that investigated aggregation phenomena in a polydisperse colloidal dispersion of ferromagnetic nanoparticles. All these works suppose that chainlike clusters with various shapes are formed along magnetic field direction. These shapes are influenced by the magnitude of both particle-particle and particle-field interactions, which is characterized by the coupling constant. Large values of coupling constant mean agglomeration of particles in larger structures – clusters, chains. In this paper the authors study the influence of temperature on the anisotropy of the acoustic attenuation in magnetic fluids based on transformer oil MOGUL. The observed results are analyzed and discussed.

2. Experimental details

The subject of the study was magnetic fluid based on transformer oil MOGUL. The structure of magnetic fluid was investigated by acoustic spectroscopy. The magnetic fluid used in experiments consisted of magnetite nanoparticles ($\text{FeO}\cdot\text{Fe}_2\text{O}_3$) with the mean diameter $d = 9.8$ nm ($\sigma = 0.28$ nm), coated with oleic acid as a surfactant dispersed in transformer oil MOGUL. The basic properties of this magnetic fluid, such as the density,

* ¹Jozef Kudelcik, ¹Peter Bury, ²Peter Kopcansky, ²Milan Timko

¹Department of Physics, Faculty of Electrical Engineering, University of Zilina, Slovakia

E-mail: kudelcik@fyzika.uniza.sk

²Department of Magnetism, IEP SAS, Kosice, Slovakia

saturation magnetization and volume fraction were equal to 0.89 g/cm³ and 3.39 mT for 1% magnetic fluid and 0.95 g/cm³ and 6.17 mT for 2% magnetic fluid.

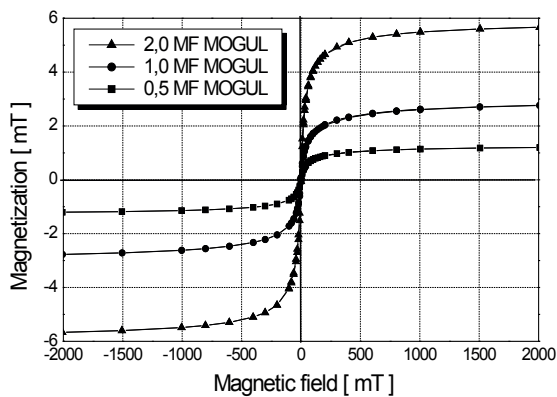


Fig. 1 Magnetization curves for various concentrations of magnetic nanoparticles in magnetic fluid based on transformer oil MOGUL

Volume concentrations, average diameter and standard deviation of magnetic particles were determined from vibrating sample magnetometer measurements. The dependences of magnetic moment of samples on magnetic field were measured in the range of -2T to 2T at room temperature (25 °C) (Fig. 1). The acoustic wave attenuation was measured at the frequency 12.65 MHz as a function of magnetic field and for different temperature. The experimental arrangement of acoustic spectrometer has been already described [12 and 13]. The acoustic velocity was measured using the double distance between transducers (18 mm) and time between the first two selected adjacent echoes.

3. Results and discussion

The attenuation of acoustic wave in magnetic fluid depends on the magnetic field intensity, the rate of its changes and the temperature [13]. The initial increase of acoustic attenuation with increasing magnetic field is connected with the aggregation of magnetic nanoparticles and following clusters creation due to the presence of magnetic field and particle-field interaction. This process can continue also at decreasing magnetic field and the development of attenuation shows then hysteresis which strongly depends also on the temperature. At lower temperature the creation of clusters is more effective because Brown thermal motion is not so effective to destroy the clusters [6 and 10]. The thermal motion increases with increasing temperature that results

in decrease of number of clusters and their size and, subsequently, also in decrease of acoustic attenuation.

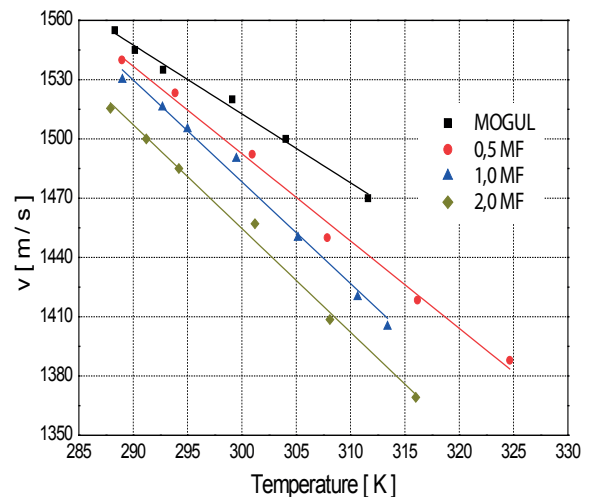
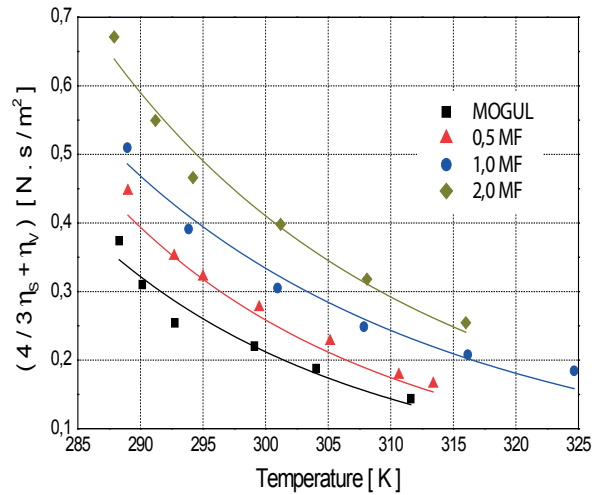


Fig. 2 The temperature dependences of term $(4/3 \eta_s + \eta_v)$ (a) and velocity (b) for various concentrations of magnetic nanoparticles in magnetic fluid based on MOGUL

More information about existence, size and density of cluster can be obtained from the analysis of the dependence of the acoustic attenuation on the angle ϕ between wave vector k and direction of magnetic field B (anisotropy). These parameters can be calculated using Taketomi theory [14] that unlike another, Shliomis and Mond theory [15] often used to describe the anisotropy of acoustic attenuation, is able to explain measured dependences in magnetic fluids based on transformer oil [13].

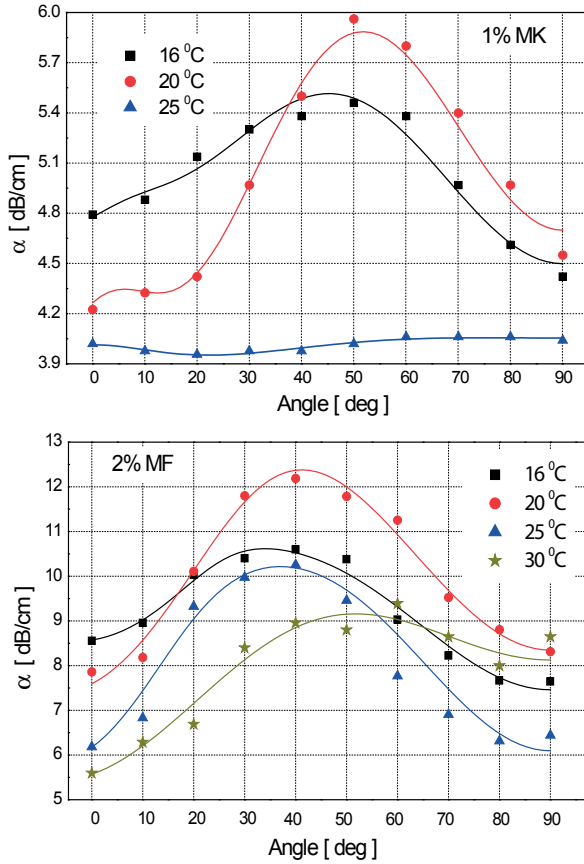


Fig. 3 Anisotropy measurement of the acoustic attenuation ($B = 200$ mT) in the 1% (a) and 2% (b) MF based on MOGUL measured at various temperatures (■ 16 °C, ● 20 °C, ▲ 25 °C) including the sum of the components $\alpha_{rot} + \alpha_{tr}$ of the Taketomi functions [14] (full lines)

Magnetic fluid in the presence of external magnetic field consists of individual magnetic nanoparticles, long chains, clusters and their various combinations which in interaction with acoustic wave cause its additional absorption. In general, the acoustic attenuation is the sum of rotation and translation part [14]

$$\alpha = \alpha_{rot} + \alpha_{tr}. \quad (1)$$

The rotating mechanism affecting the part of acoustic attenuation is described as

$$\alpha_{rot} = \frac{2\pi^2 f^2}{\rho_0 c^3} \left(\left(\frac{4}{3} \eta_s + \eta_v \right) + 2\alpha_5 \cos^2 \varphi + \alpha_1 \cos^4 \varphi \right), \quad (2)$$

where ρ_0 is the density of the carrier fluid, c is the velocity of the acoustic wave of the frequency f , $(\frac{4}{3} \eta_s + \eta_v)$ is viscosity, α_1, α_5 are the Leslie coefficients appearing in the theory of liquid

crystals. Three types of forces are assumed in his theory. The first one is a recovering force that acts on the cluster toward to original point and makes then periodic motion. The second force is the frictional force, that decelerates the moving cluster and particles in viscose liquid. From the acoustic theory of fluids, the last force is derived - the force of the pressure. From the solution of the equation of motion for a cluster and expression of the dissipative energy the translational acoustic attenuation was derived in the form

$$\alpha_{tr} = \frac{3\pi\eta_0 a \omega^3 \rho_0 V N (6\pi\eta_0 a + \rho_0 V \omega) / (kc^2)}{(\sin \varphi - \rho_m V \omega^2 / k)^2 + (6\pi\eta_0 a \omega / k)^2}, \quad (3)$$

where k is the constant of recovering force, a is the radius of clusters and N is the density of clusters.

The second and the third terms in Eq. 2 are non zero only in presence of external magnetic field. In a magnetic fluid not subjected to an external magnetic field, the acoustic attenuation, α_{rot} , is firstly related to the dynamic viscosity, η_s , and volume viscosity, η_v [9]. The temperature dependence of the term $(\frac{4}{3} \eta_s + \eta_v)$ enables the fit with the Arrhenius function

$$\left(\frac{4}{3} \eta_s + \eta_v \right) = f(T) = A \exp \left\{ \frac{B}{T} \right\}, \quad (4)$$

where A and B are coefficients dependent on the properties of magnetic fluids. These coefficients are summarized in Table 1. As follows from Fig. 2a illustrating the temperature dependence of this term, the coefficient of viscosity decreases with increasing temperature. The coefficient increases also with the concentration of magnetic nanoparticles in magnetic fluid. From Fig. 2b it can be seen that the velocity of acoustic wave decreases with increasing temperature that corresponds to the increasing density of magnetic fluid. In this case the velocity is smaller for higher concentration of magnetic nanoparticles in magnetic liquid. The velocity of acoustic wave in investigated magnetic fluid based on transformer oil can be fitted by function

$$c = c_0 (1 - \beta \Delta T), \quad (5)$$

where c_0 is the velocity at temperature 273K and β is the coefficient of the temperature change of the velocity. Calculated values are also summarized in Table 1.

Parameters of the temperature dependences of term $(\frac{4}{3} \eta_s + \eta_v)$ and velocity for various concentrations of magnetic nanoparticles in magnetic fluid based on MOGUL

Table 1

X% MF	c_0 [m/s]	β [mK ⁻¹]	A [10 ⁻⁶]	B [K]
MOGUL	1607	2.2	1.2	3628
0.5%	1612	2.7	1.3	3658
1.0%	1617	3.2	1.0	2944
2.0%	1596	3.3	1.0	3160

Type values of parameters described 1% and 2% magnetic fluids based on the MOGUL obtained from the fit of measured anisotropy data using Taketomi functions

Table 2

X% MF MOGUL	1%			2%			
	16 °C	20 °C	25 °C	15 °C	20 °C	25 °C	30 °C
Temperature	16 °C	20 °C	25 °C	15 °C	20 °C	25 °C	30 °C
$4/3\eta_s+\eta_v$ [N.s.m ²]	0.38	0.35	0.32	0.58	0.45	0.39	0.36
α_s [N.s.m ²]	0.18	0.25	0.00022	0.33	0.40	0.48	0.16
α_1 [N.s.m ²]	-0.39	-0.73	-0.02249	-0.56	-0.76	-1.03	-0.6
k [N.m ⁻¹]	11.73	7.88	2.52	66	42	37	10
a [nm]	93	62	19	290	265	217	135
$10^{17} N$ [m ⁻³]	9.8	102	313	0.5	2	3.1	17.3
$V \times N$ [10 ⁻³]	0.42	1.27	0.11	0.64	1.95	1.66	2.22

The anisotropy of acoustic attenuation measured at magnetic field 200 mT for two different concentrations of magnetic nanoparticles in magnetic fluid is shown in Fig. 3. The solid lines represent the theoretical fit of experimental data using Taketomi functions (Eqs. 2, 3). The measured anisotropy shows curves with the maximum value whose position depends on the concentration and temperature: for 1% MF MOGUL: 16 °C - 45°, 20 °C - 50°, 25 °C - no evident maximum; for 2% MF MOGUL: 15 °C - 40°, 20 °C - 40°, 25 °C - 40°, 30 °C - 55°. Using rotation $\alpha_{rot}(\phi)$ and translational $\alpha_{tr}(\phi)$ part of Taketomi function next parameters can be determined: ($4/3 \eta_s + \eta_v$) (viscosity), α_s , α_1 , k (constant of recovering force), a (radius of clusters) and N (density of clusters). These parameters obtained from the fit of measured data in Microcal Origin are summarized in Table 2.

Figure 4 shows both anisotropy of the acoustic wave absorption in 1% and 2% magnetic fluid measured at 20 °C as well as the individual components α_{rot} , α_{tr} obtained from Taketomi functions (Eqs. 1, 3). As follows from the dependence $\alpha_{rot}(\phi)$, the curve has a maximum value whose position depends on concentration: 1% - 55°, 2% - 50° and temperature, too. On the other hand, the second component of the acoustic attenuation, $\alpha_{tr}(\phi)$, decreases monotonously with increasing angle ϕ for lower concentration (1% MF), but at higher concentration (2% MF) reaches the maximum value around 40°.

The volume concentration of all clusters ($V \times N$) can be determined also from the parameters of a and N . This value implies which part of the magnetic grains (domains) is included in the clusters, while other part is free in the carrier fluid. As it can be seen from the volume ($V \times N$) more than 4,2% of all magnetite particles are in clusters.

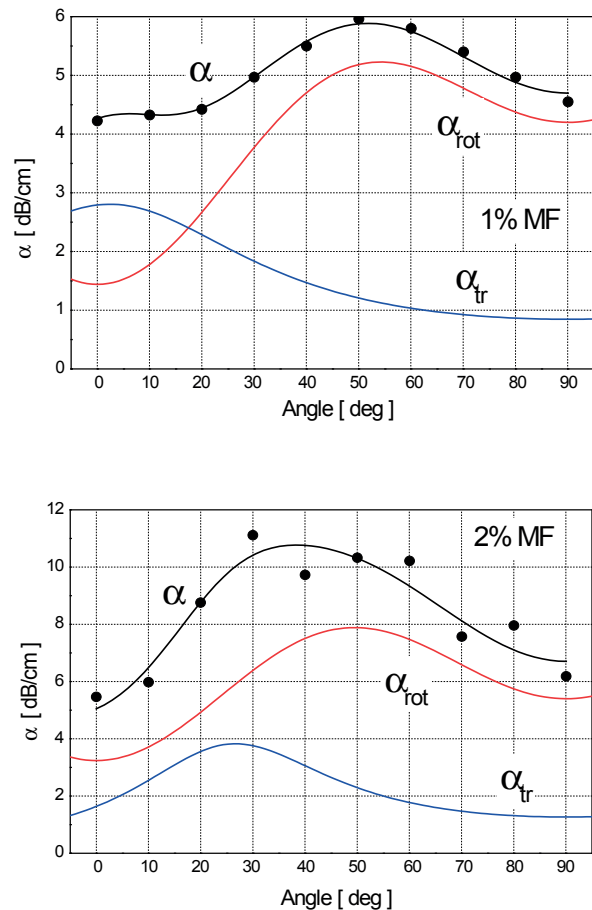


Fig. 4 Development of rotation and translation part of acoustic attenuation and their sum measured in the 1% and 2% MF based on MOGUL for temperature 20 °C

The measurement of the acoustic attenuation as a function of the angle φ between the direction of propagation represented by k and that of the magnetic field B (anisotropy) for magnetic fluids when the majority of the particles are involved in the cluster structures, confirmed the contribution of two components of the acoustic attenuation α_{rot} , α_{tr} related to the rotational and translational degrees of freedom, respectively. The translational and rotation parts of acoustic attenuation express also what part of acoustic wave energy losses is due to the translation and/or rotational movement of the clusters. So that it is possible also to estimate the percentage contribution of acoustic attenuation to the individual kinds of cluster motion.

It is known that the interaction between the external magnetic field and the magnetic moment of nanoparticles in magnetic fluids leads to the aggregation of nanoparticles to new structures. These structures enlarge with the increasing magnetic field and this process has the influence on the value of the acoustic attenuation, so that the acoustic attenuation increases, too. The acoustic attenuation initially increases with increasing magnetic field due to the starting process of aggregation of magnetic particles to the cluster and later to chains (structures as long as tens of nanometers, see Table 2). However, following progress at higher magnetic field can be different depending on the structure changes caused by developing of cluster shape in individual cases.

The complex viscosity as a function of temperature was determined from the measurement of acoustic attenuation for various concentrations and temperature. It can be seen that viscosity decreases with temperature and increases with concentration. The similar results were observed from the velocity measurements. The velocity decrease with the concentration is caused by higher numbers of nanoparticles in magnetic fluid and consequently its higher density.

The important feature of studied magnetic liquid is the anisotropy of acoustic attenuation, which was investigated in the temperature range of 16 - 30 °C. The obtained results indicate the significant effect of temperature on the acoustic attenuation. The measurements of the anisotropy of acoustic attenuation in the case of 1% MF MOGUL at temperatures 16 and 20 °C show large changes connected with the process of chains orientation and its rotation in the direction of magnetic field. At higher temperature (>24 °C) the acoustic attenuation is almost independent on magnetic field. There are also other parameters discovered by acoustic attenuation: the radius of clusters and the length of their chains. These parameters are strong function of temperature. Its mean value changes from 93 nm to 19 nm corresponding to the temperature increase from 16 °C to 25 °C.

The measurement of the anisotropy of acoustic attenuation for 2% MF MOGUL even indicates stronger effect as that for 1%

concentration, when the radius of clusters at the same magnetic field and temperature is much higher. For this concentration the anisotropy of acoustic attenuation also at temperature 25 °C shows large changes connected with the process of chains orientation and its rotation concerning the direction of magnetic field. The anisotropy measured at 30 °C still has maximum although less expressive. Very important fact is that the values of cluster radius at lower temperature (16 °C) can be larger than 250 nm, comparing with the value 93 nm for 1% MF. This is explanation also for dominant effect of rotation part on acoustic attenuation relative to translation part. This big difference in cluster radius indicates also that clusters in this type of MF look like separate spheres and they do not create long chains.

At higher temperature the thermal motion increases resulting in decrease of numbers of clusters and mainly their radius and volume concentration. The both smaller numbers of clusters and their smaller size induce the smaller influence on the acoustic attenuation. The changes in the clusters size affect also the magnitude of the rotation and translation components of the acoustic attenuation.

4. Conclusions

The influence of both magnetic field and temperature on the structures of investigated magnetic fluids based on the transformer oil MOGUL was observed using acoustic spectroscopy. The anisotropy of the acoustic attenuation in the presence of external magnetic field of the value 200 mT for two different concentrations was measured and analyzed. The effect of temperature on the cluster radius and their configuration considering the direction of magnetic field was confirmed. The study of the anisotropy showed also the important role of rotational and translational motion of the clusters on the acoustic attenuation and the fact that the concentration of magnetic nanoparticles in transformer oil MOGUL has influence on the size of clusters and their arrangement. Using Taketomi theory the radius of clusters, their density and viscous term as a function of temperature were determined.

Acknowledgement

This work was supported by VEGA projects 1/0624/13 and 2/0045/13. The authors wish to thank also for the support to the R&D operational program Centrum of excellence of power electronics systems and materials for their components, No. OPVaV-2008/2.1/01-SORO, ITMS 2622012046 funded by European Community.

References

- [1] SEGAL, V., RAJ, K.: An Investigation of Power Transformer Cooling with Magnetic Fluids, *Indian J. Eng. Mater. Sci.* 5 (6), 1998, 416.
- [2] ODENBACH, S.: Ferrofluids - magnetically Controlled Suspensions, *Colloids and Surfaces A: Phys. Eng. Aspects*, 217, 2003, 171.
- [3] KUDELCEK, J., BURY, P., ZAVISOVA, V., TIMKO, M., KOPCANSKY, P.: The Dielectric Breakdown Of Magnetic Fluids, *Communications - Scientific Letters of the University of Zilina*, vol. 12, Nr. 2. 2010, 34-37.
- [4] KUDELCEK, J., BURY, P., KOPCANSKY, P., TIMKO, M.: Dielectric Breakdown in Mineral Oil ITO 100 Based Magnetic Fluid, *Physics Procedia* 9, 2010, 78-81.
- [5] YUE-FAN, DU, YU-ZHEN, LV., JIAN-QUAN, Z. XIAO-XIN, LI, CHENG-RONG, LI: *Breakdown Properties of Transformer Oil-based TiO₂ Nanofluid Electrical Insulation and Dielectric Phenomena (CEIDP)*, 2010 Annual Report Conference on Electrical Insulation and Dielectric Phenomena 2010 <http://dx.doi.org/10.1109/CEIDP.2010.5724082>.
- [6] MENDELEV, V. S., IVANOV, A. O.: Ferrofluid Aggregation in Chains under the Influence of a Magnetic Field, *Phys. Rev.* 70, 2004, 051502
- [7] ROZYNEK, Z. et al: Structuring from Nanoparticles in Oil-based Ferrofluids, *Eur. Phys. J. E* 2011, 34: 28 DOI 10.1140/epje/i2011-11028-5
- [8] SINGH, V., BANERJEE, V., SHARMA, M.: *Dynamics of Magnetic Nanoparticle Suspensions*, *J. Phys. D: Appl. Phys.* 42, 2009, 245006
- [9] SKUMIEL, A.: The Effect of Temperature on the Anisotropy of Ultrasound Attenuation in a Ferrofluid, *J. Phys. D: Appl. Phys.* 37, 2004, 3073.
- [10] AOSHIMA, M., SATOH, A.: Two-dimensional Monte Carlo Simulations of a Colloidal Dispersion Composed of Polydisperse Ferromagnetic Particles in an Applied Magnetic Field, *J. of Colloid and Interface Science* 288, 2005, 475-488.
- [11] SATOH, A., COVERDALE, G. N., CHANTRELL, R. W.: Stokesian Dynamics Simulations of Ferromagnetic Colloidal Dispersions Subjected to a Sinusoidal Shear Flow, *J. of Colloid and Interface Science* 231, vol. 2, 2000, 238-246
- [12] KUDELCEK, J., BURY, P., DRGA, J., KOPCANSKY, P., ZAVISOVA, V., TIMKO, M.: Temperature Effect on The Structure of Transformer Oil Based Magnetic Fluids Using Acoustic Spectroscopy, *Acta Physica Polonica A*, 2012, vol. 121, No. 5-6, ISSN 0587-4246 1169-1171
- [13] KUDELCEK, J., BURY, P., DRGA, J., KOPCANSKY, P., ZAVISOVA, V., TIMKO, M.: Structure of Transformer Oil-based Magnetic Fluids Studied using Acoustic Spectroscopy, *J. of Magnetism and Magnetic Materials* 326, 2013, 75-80.
- [14] TAKETOMI, S.: The Anisotropy of the Sound Attenuation in Magnetic Fluid under an External Magnetic Field, *J. Phys. Soc. Japan* 55, 1986, 838.
- [15] SHLIOMIS, M., MOND, M.: Ultrasound Attenuation in Ferrofluids, *Phys. Rev. Lett.* 101, 2008, 074505.