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Comparison of the Change of Acoustic Attenuation and Anisotropy in Magnetic Fluids Based on Transformer Oils

J. Kúdelčík^{a,*}, Š. Hardoň^a, P. Bury^a, M. Timko^b and P. Kopčanský^b

 $^a\mathrm{Department}$ of Physics, Faculty of Electrical Engineering and Information Technology,

University of Žilina, Univerzitná 1, 010 26 Žilina, Slovakia

^bInstitute of Experimental Physics, SAS, Watsonova 47, 040 01 Košice, Slovakia

In the application of a magnetic field to a magnetic fluid, magnetic nanoparticles are created with various structures. In this contribution, we compare the structural changes in three types of transformer oil-based (TECH-NOL, MOGUL, and MOL) magnetic fluids upon different developments of magnetic fields and temperatures by the acoustic spectroscopy. The change of the acoustic attenuation was slow (> 10 min) or fast (< 4 min) depending on the step change of the magnetic field. After switching off the magnetic field, a fast decrease of the acoustic attenuation was observed only for MOL-based magnetic fluid. For linear changes of the magnetic field, the acoustic attenuation increased in steps and a hysteresis effect was observed. The effect of anisotropy of the acoustic attenuation was observed and analysed.

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1. Introduction

At present, there are various magnetic fluids (MFs) based on transformer oil [1–4]. Their properties can be studied using several methods: dielectric [5, 6], acoustic [2, 7], electrical [3, 4], etc. The acoustic spectroscopy has confirmed that the change in acoustic attenuation (α) in MFs depended on the magnitude of the magnetic field, rate of its change, and also on temperature [2, 7]. The anisotropy measurements are also very important whereby theoretical models can be used to determine type and size of nanoparticle structures [3, 7, 8].

2. Experimental parameters

The experiment used three types of MFs based on transformer oils: TECHNOL, MOGUL, and MOL. All MFs contained the same type of nanoparticles: $FeO \cdot Fe_2O_3$ with a mean diameter of around 10 nm and coated with oleic acid as a surfactant. In all cases, we used 1% volume concentration of magnetic nanoparticles in the given oil. Their densities, viscosities, acoustic attenuation of given MF/clean oil without magnetic field and magnetizations were (882, 890, 910) kg/m³, (0.24,0.35, 0.38) N s/m², (3.9/3.2, 3.3/2.4, 5.2/4.7) dB/cm, and (4.7, 3.4, 3.3) mT, respectively at temperature of 25 °C. MF was placed in the thermostated closed measuring cell (Fig. 1, point 3) with two piezoelectric transducers (Fig. 1, point 4) with frequency of 11.5 MHz [7]. From the amplitude ratio of the acoustic pulses, the acoustic attenuation was determined. The analysis was performed on the received signals in the time domain, and



Fig. 1. Experimental arrangement of thermostated measuring cell in electromagnet at (a) step and linear change of the magnetic field, and (b) anisotropy measurement. Experimental components: part 1 — electromagnet, part 2 — temperature stabilization block, part 3 — measuring cell, part 4 — piezoelectric transducers, \boldsymbol{B} — the direction of the magnetic field, \boldsymbol{k} — the direction of the acoustic pulse, α — the angle between \boldsymbol{B} and \boldsymbol{k}).

the attenuation coefficient α was obtained from the following expression: $\alpha = \frac{1}{d} \ln \left(\frac{A_1}{A_2}\right)$, where A_1 , A_2 are the amplitudes of the signal transmitted through the sample, and d = 18 mm was the difference in the acoustic paths traveled by the signal.

3. The step-change of the magnetic field

The first measurement was a response of MFs to the step change of the magnetic field at various temperatures (Fig. 2). The magnetic field \boldsymbol{B} was parallel to the acoustic wave vector \boldsymbol{k} (Fig. 1a). At first, the magnetic field was zero for 10 min and then it was switched on to 200 mT. This value was unchanged during a certain period of time, and then it was switched off to 0 mT. During the application of the magnetic field, a gradual change of the acoustic attenuation ($\Delta \alpha$) to a steady value

^{*}corresponding author; e-mail: kudelcik@gmail.com



Fig. 2. Experimental data of changes in the acoustic attenuation for the step change of the magnetic field to value 200 mT measured at various temperatures and types of MF: (a) MF TECHNOL, (b) MF MOGUL [9], (c) MF MOL, and (d) comparing the given MFs at 25 °C.

was observed. No change of $\Delta \alpha$ in clean carried fluids was observed due to the application of the magnetic field. For MF MOL stable values were observed within 20 min and for MF TECHNOL and MF MOGUL it took a longer time. The observed changes in $\Delta \alpha$ were caused by structural changes in the MFs, which means that magnetic nanoparticles created new structures [7, 9].

On the basis of previous works at the given magnetic field, we can assume the existence of short thick chain structures or clusters. The different developments of $\Delta \alpha$ clearly confirmed that the processes leading to the formation of structures were dependent also on the type of carrier fluid. Independent, of the type of MF, the change of acoustic attenuation $(\Delta \alpha)$ was smaller at higher temperatures. The maximal values of $\Delta \alpha$ were similar for all MFs at temperature 25 °C (Fig. 2d). Switching off the magnetic field should be associated with the disintegration of the structures. A decrease in the acoustic attenuation of more than 20 min (MF TECHNOL, MF MOGUL) means that structures had long lifetime, maybe due to interparticle forces. In practice, this effect is generally not positive because larger structures can negatively affect the electrical insulation properties [10]. Only in case of MF MOL, a quick decrease of α to the initial values after the magnetic field was switched off, was observed.

4. Linear change of the magnetic field

Figure 3 presents $\Delta \alpha$ in studied MFs for the linear change of the magnetic field from 0 mT to 200 mT and back (3.3 mT/min) at various temperatures. In all cases, we can observe an increase in acoustic attenuation with the magnetic field. This increase corresponded to a stepby-step increase in the size and number of nanoparticle structures. This phenomenon was observed also by other authors [7, 9]. The change of $\Delta \alpha$ was higher for



Fig. 3. Changes of the acoustic attenuation on the linear change of the magnetic field to 200 mT in various MF measured at 25 °C. The magnetic field increased and decreased at a constant rate of 3.3 mT/min.

MF TECHNOL as for MF MOGUL and MF MOL. Interestingly, a hysteresis loop was observed for the decrease of the magnetic field. This loop was a consequence of the processes of the formation of structures that occurred even at a decreasing magnetic field. The decrease of $\Delta \alpha$ was minimal for MF TECHNOL and MF MOGUL because the lifetime of structures was longer than the time of decrease. In the case of MF MOL the lifetime was smaller than 10 min (Fig. 2c) so the acoustic attenuation returns to the initial value at 0 mT. Independent of the type of MF for increasing temperature, there was almost no change in $\Delta \alpha$ with the linear change in the magnetic field.

5. Anisotropy of MF

The anisotropy measurements of the acoustic attenuation in two MFs for three different temperatures are shown in Fig. 4. For these measurements, the magnetic field was firstly switched on to 200 mT ($\boldsymbol{B} \parallel \boldsymbol{k}$) and remained constant during the whole time. After 60 min the first value of α at angle 0° ($\boldsymbol{B} \parallel \boldsymbol{k}$) was measured. Then the angle between \boldsymbol{B} and \boldsymbol{k} was changed to 10° (Fig. 1b) and the next value of α was measured after stabilization time of 15 min [7, 9]. For the next angles, we used the same procedure. The full lines correspond to the theoretical fit of experimental data using Taketomi functions [8] parameters of which are summarized in Table I.

At lower temperatures, the acoustic attenuation had local maxima at angles — MK TECHNOL, i.e., $(40^{\circ} - 20^{\circ}\text{C}, 50^{\circ} - 25^{\circ}\text{C})$, and at 50° MK MOGUL $(16^{\circ}\text{C}, 20^{\circ}\text{C})$. At higher temperatures, the change in acoustic attenuation was almost independent of the direction of the magnetic field. Based on the works [7, 9] we can say that the observed maxima are the result of the presence of bigger nanoparticle structures, i.e., clusters. The rotational motion of these clusters influences the increase of the rotational component of the acoustic attenu-



Fig. 4. Anisotropy of the acoustic attenuation at 200 mT in MF TECHNOL [7] and MF MOGUL [9] measured at various temperatures including the sum of the Taketomi functions [8] (full lines).

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Parameters described MF based on the TECHNOL and MOGUL obtained from the fit of measured anisotropy data using Taketomi functions $(4/3 \eta_S + \eta_V)$ — viscosity [N s/m²], k — binding constant [N/m], a — cluster radius [nm], $V \times N$ — average volume of clusters times their concentration of all structures [%] [7, 9]

| | MF TECHNOL | | | MF MOGUL | | |
|----------------------|------------|------|------|----------|------|------|
| $T [^{\circ}C]$ | 20 | 25 | 30 | 16 | 20 | 25 |
| $4/3\eta_S + \eta_V$ | 0.28 | 0.24 | 0.23 | 0.38 | 0.35 | 0.32 |
| k | 2.3 | 0.55 | 0.24 | 11.73 | 7.88 | 2.52 |
| a | 49.7 | 34 | 24 | 93 | 62 | 19 |
| $V \times N$ | 0.33 | 0.11 | 0.08 | 0.42 | 1.27 | 0.11 |

ation [8], which results as the observed maximum. Their existence was confirmed by parameters (Table I) obtained from the Taketomi fit of anisotropy measurements. From the viscosity term, namely $\frac{4}{3}\eta_S + \eta_V$, where η_S is shear viscosity and η_V is volume viscosity, we conclude that it decreased with temperature for all studies of transformer oils. The different behaviors of investigated MFs in the magnetic field were also affected by their different viscosities. If the viscosity term is smaller, the movement of structures is easier. Therefore the thermal Brown motion was more effective and corresponding values of acoustic attenuation changes were smaller. The next important fact was that the mean value of nanoparticle structures or clusters decreased from 93 nm to 19 nm for MF MOGUL and from 50 nm to 24 nm for MF TECHNOL with temperature. At low temperatures the cluster consists of several tens of nanoparticles and at higher temperatures, there are only about 2–3 nanoparticles. In that case, we can say that nanoparticles structures look like short chains. These small structures have almost no rotational motion, so they have minimal effect on the change on the acoustic attenuation, i.e., no anisotropy effect.

6. Conclusions

Acoustic spectroscopy was used to study the influence of magnetic field on the structural changes in magnetic fluids based on various types of transformer oils. These structural changes have emerged as a change in the acoustic attenuation due to the application of the magnetic field. The different values of the acoustic attenuations were caused by different sizes and numbers of nanoparticle structures and their lifetimes. In the experiment, measurements showed that the time-change of acoustic attenuation was also dependent on the types of transformer oils and their viscosities. The fast changes in the acoustic attenuation (structural change) were observed only in MF MOL. In all types of transformer oils at temperatures higher than 30 °C the magnetic field had minimal effect on the change of acoustic attenuation.

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