STRUCTURAL PROPERTIES OF MAGNETIC FLUID STUDIED BY ACOUSTIC SPECTROSCOPY

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1. Introduction

Magnetic fluids have found wide application in technology, medicine and other areas to improve the properties of various materials. By adding the magnetic nanoparticles to the transformer oil their properties interesting in power electronic technology can be improved [1, 2]. The transformer oil-based magnetic fluids can have better insulating and thermal properties. The aggregation effects of magnetic nanoparticles in external magnetic field strongly influenced on electric breakdown [3, 4].

Acoustic wave propagation in magnetic fluids was studied by several authors both theoretically and experimentally [5-9]. They also showed that an external magnetic field has big effect on the change of acoustic attenuation. This is caused by aggregation of the nanoparticles of magnetic fluid along magnetic field direction into oligomers, chains or clusters [7, 9-12]. These shapes depend on both particle-particle interactions represented by the coupling constant and particle-field interactions, which are also important in computer simulations [13, 14]. New information can be also obtained from the study of the anisotropy of acoustic attenuation coefficient as the function of the angle φ between the magnetic field direction and the propagation direction of acoustic wave. Two kinds of motions of structures from ferrous colloidal nanoparticles in the fluid can be then recognized. The first one is the rotation motion and the second the translation motion, which were analyzed by the model of a vibrating sphere in the viscous fluid [10, 15, 16].

In this paper the interference of magnetic field, time and temperature on the structure of magnetic fluids based on transformer oils MOGUL and TECHNOL with same concentration but various diameters of nanoparticles are presented. The structure changes are studied by acoustic spectroscopy, observed results for both kinds of magnetic fluids are discussed and compared.

2. Experimental results

Transformer oils MOGUL and TECHNOL were used as carrier liquids for the preparation of magnetic fluids for the investigation by acoustic method. The magnetic fluids used in experiments consisted of magnetite nanoparticles (FeO×Fe₂O₃) with the mean diameter d = 7.9 nm and d = 11.1 nm (σ = 0.28 nm), respectively, coated with oleic acid as a surfactant and dispersed in individual transformer oil. The basic properties of investigated magnetic fluids, such as the density, saturation magnetization and volume fraction were equal to 0.89 g/cm³, 3.39 mT for 1% magnetic fluid based on MOGUL and 0.882 g/cm³ and 4,73 mT for 1% magnetic fluid (MF) based on TECHNOL. The speed of ultrasonic wave and viscosity term as a function of temperature for MFs based on given transformer oil are presented in Fig. 1. The ultrasonic wave absorption was measured for the frequency 12.65 MHz, as a function of magnetic field and for different temperature. The block diagram of the experimental arrangement was used as in the work [10, 11].



Fig.1:The velocity (a) and temperature dependences of term $(4/3 \eta S + \eta V)$ (b) for various concentrations of magnetic nanoparticles in magnetic fluid based on MOGUL and TECHNOL.

Figure 2 shows the changes of acoustic attenuation when the magnetic field is linearly increased (3,3 mT per minute) to the maximal value 300 mT for MF MOGUL based on and to the maximal value 200 mT for MF based on TECHNOL. At the first sight it can be seen that the observed results for both MFs have different development of change of acoustic attenuation. The results indicate a significant effect of magnetic field and temperature on the acoustic attenuation. At the lowest temperature 15 0 C, the change of the acoustic attenuation with magnetic field is very pronounced. There is the significant increase of attenuation with increasing magnetic field. At higher temperatures, 20 0 C and 25 0 C, the developments of the change of acoustic attenuation are only slight function of magnetic field. At these temperatures large structures of nanoparticles cannot be created and nanoparticles are practically free or create small oligomers.



Fig.2: The dependence of acoustic attenuation changes on the external magnetic field for MF MOGUL (a) and MF TECHNOL (b) measured at various temperatures. Measurement at the temperature 20 °C in case (a) is shifted +0.2 dB/cm for better resolution.

The measurement at the lowest temperature (15 °C, Fig. 2b) shows the largest changes originated from the process of particle agglomeration, chains aggregation and clusters creation. At this temperature the creation of clusters is more effective because Brown thermal motion is not so effective to destroy them [5]. At higher temperatures the thermal motion increases resulting in decrease of both number and size of clusters. The smaller numbers of clusters induce the smaller change of acoustic attenuation. At temperature 20 °C and 25 °C the changes of acoustic attenuation are smaller. At the temperature 30 °C the structure of clusters almost disappears and the majority of domains are practically free. The influence of magnetic field on the acoustic attenuation happens small and as follows from our results the majority of the particles are not involved in the cluster structures [14, 17].

More information about arrangement of nanoparticles into structures, their size and density can be obtained from the analysis of the dependence of the acoustic attenuation on the angle φ between the acoustic wave vector k and the direction of magnetic field B (anisotropy). These parameters can be calculated using the suitable theory [16] that takes into account both translational and rotational movement of nanoparticles formations.



Fig.3:Anisotropy measurement of the acoustic wave attenuation (B = 200 mT)for (a)MFMOGUL and (b)MF TECHNOL.

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The results of anisotropy of acoustic attenuation measured at magnetic field 200 mT for four different temperatures of MFs are shown in Fig. 3. The measurements were made in next steps: at the begin, the jump change of magnetic field was from value 0 mT to 200 mT and next 120 minutes we waited for steady-state. Then at 200 mT, the angle between direction of the magnetic field and the wave vector of acoustic wave was changed by 10 Deg to 90 Deg. The measurement of the acoustic attenuation at new angle was done after 15 minutes of stabilization. The solid lines represent the theoretical fit of experimental data using theoretical functions [10, 16], where main parameters (k - constant of recovering force, a - radius of clusters, N - density of clusters and volume concentration of all clusters $V \times N$) are presented in Table 1 [10, 11, 12]. It can be seen that the changes of acoustics attenuation for

MF TECHNOL are higher as MF MOGUL. The measured anisotropy has maximum value whose position depends on temperature, however only for lower temperatures, for MF MOGUL: $15 \degree C - 40\degree$; for MF TECHNOL: $20 \degree C - 45\degree$, $25 \degree C - 50\degree$.

	MF TECHNOL			MF MOGUL		
Temperature	20 °C	25 °C	30 °C	15 ⁰ C	20 °C	25 °C
$4/3\eta_{\rm S}+\eta_{\rm V}$	0.29	0.24	0.23	0.35	0.32	0.24
$k[\text{ N.m}^{-1}]$	2.3	0.55	0.24	1.5	1.1	0.5
<i>a</i> [nm]	50	34	24	71	20	16
10 ⁻¹⁷ N[m ⁻³]	255	277	560	22.7	507	549
VxN [%]	0.33	0.11	0.08	0.34	0.17	0.09

Tab. 1.Parameters describing magnetic fluids based on the TECHNOL and MOGUL obtained from the fit of measured anisotropy data.

3. Discussion

The interaction between the external magnetic field and the magnetic moment of the nanoparticle in magnetic fluids leads to the aggregation of nanoparticles to new structures [6, 8, 10, 11]. These structures enlarge with the magnetic field and they have the influence on the value of the acoustic attenuation. Its value increases with increasing magnetic field and at decrease of magnetic field the speed of decrease dependent on properties of magnetic fluids [5, 12]. This effect can be explained by several parameters. One of is the time constant of creation of higher structures of nanoparticles or other are temperature and viscosity of given magnetic fluids.

The slow increase of magnetic field effects on the structural changes and by this means the acoustic attenuation (Fig. 2). There is enough time for gradual creation of structures like: dimers, trimers and higher oligomers. On the basic of experimental measurement we could say that for MF TECHNOL and at higher magnetic field (> 200 mT) in MF MOGUL long thin chains or clusters can be created. However, these processes are different in dependence on the size of nanoparticles, temperature and time for the creation of these structures from magnetic nanoparticles which are about several tens of minutes [10]. The change of acoustic attenuation in the case of MF MOGUL was the biggest at the temperature 15°C. At higher temperatures the changes of acoustic attenuation were also independent on magnetic field. For MF TECHOL the situation is other, because until from temperature 30°C there is no change of the acoustic coefficient. At higher temperature for both type of MF the change of acoustics attenuation is much smaller, because there is more significant effect of thermal motion. At these temperatures the numbers of the larger structures are less and are also smaller, so they have smaller effect on the acoustic attenuation.

The information about size of structures was gained from the anisotropy of acoustic attenuation measured at various temperatures. These measurements show that only at the lowest temperature for MF MOGUL and for 20 and 25°C for MF TECHNOL bigger structures can exist. This big difference in temperature range is caused by size of nanoparticles in studied MFs. For smaller nanoparticles thermal Brown motion (speed) and viscosity are higher so at temperatures bigger than 20°C only dimer (trimer) can exist. For bigger nanoparticles (MF TECHNOL) the structures are no bigger but they have better

stability. Smaller viscosity and stable structures result in more pronounced anisotropy for this type of MF.

On the base of the calculated radius (Table 1) at the lowest temperature we can say that in the bulk in chains are more as 6 nanoparticles. So we can say that nanoparticles created clusters. At temperature 25 0C there are only 2-3 nanoparticles in the bulk, so they can create only thin chains. The both smaller number of nanoparticles in clusters or chains and thus their shorter size induce the smaller influence of the direction of magnetic field with regards to the direction of acoustic wave propagation on the acoustic attenuation.

4. Conclusion

The changes of acoustic attenuation in magnetic fluids in the external magnetic field were measured. Two kinds of magnetic fluids based on transformer oil MOGUL and TECHNOL were studies to discover the structure of nanoparticles. The magnetic field, temperature and radius of nanoparticles are important factors that influence on the structures of investigated magnetic fluids. The effect of different radius of nanoparticles in studies magnetic fluids on the values of acoustic attenuation at measured temperatures in magnetic field was observed. Using Taketomi theory and the anisotropy of the acoustic attenuation the radius of structures, their density and viscous term as a function of temperature were determined.

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