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DIELECTRIC RELAXATIONS IN A TRANSFORMER OIL-BASED MAGNETIC FLUID

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Transformer oil-based magnetic fluids are of great scientific interest due to their enhanced thermal properties and evident higher breakdown voltage if compared with pure transformer oil. To understand the paradoxical and peculiar dielectric properties of such magnetic fluids, one needs to reveal and identify all polarization processes contributing to the total polarization of the material. Even though power transformers exploiting transformer oil operate at 50 or 60 Hz frequency, it is desirable to investigate the oils dielectric properties in a broader frequency range, too. In this study, we show some selected dielectric spectra of a magnetic fluid consisting of transformer oil and magnetite nanoparticles stabilized by oleic acid. Complex dielectric permittivity spectra of the studied sample exhibit two pronounced dielectric dispersions associated with the space charge migration and interface relaxation phenomena.

Introduction. Transformer oils and oil-paper insulation are widely used in electrical power equipment, such as transformers and inductors [1, 2]. However, faults and degradations caused by thermal ageing have emerged in large numbers due to the low cooling efficiency of insulating systems. In recent years transformer oil-based magnetic fluids (ferrofluids) have attracted much attention for their great improvement of heat transfer efficiency [3–8]. In regard to the general "6°C rule" stating that the rate of aging doubles approximately every 6°C increase in temperature [9, 10], the ferrofluids constitute a promising efficient and reliable cooling medium for power transformers. Moreover, it has been found that the transformer oil-based ferrofluids (TOFF) can exhibit higher electrical breakdown field strength if compared to pure transformer oils [11]. This paradoxical finding is still not fully understood, and great effort has been made to study dielectric properties of the innovative kind of dielectrics in view of using various transformer oils, magnetic nanoparticles and surfactants.

Segal *et al.* [11] for the first time showed that the electrical breakdown field strength of TOFF containing magnetite nanoparticles is comparable or in some cases superior to that of pure transformer oil. Particularly, electric breakdown testing of magnetite TOFF has revealed that for positive streamers the breakdown voltage of the TOFF is almost twice that of the pure oil during lightning impulse tests. This result was radical in that it suggested that the presence of substantial particulate contamination in transformer oil, albeit in the form of magnetite nanoparticles could result in an improvement of the oils insulating characteristics. After the first report, several research works experimentally confirmed that paradoxical effect in TOFFs based on various transformer oils and magnetic nanoparticles [12–16]. Recently, three times higher breakdown voltage was reported for a TOFF with a particle volume fraction from 0.08% to 0.39% if compared to its base transformer oil [15]. Interestingly, along with the higher breakdown field strength, the propagation velocity of positive streamers was found to be reduced by the presence of nanoparticles by 46% [11]. This result is very important in that it indicates that the presence of magnetite nanoparticles in the oil inhibits processes leading to electrical breakdown. In other words, a slower streamer requires more time to traverse the gap between electrodes to cause breakdown. Consequently, this allows more time for the applied impulse voltage to be extinguished.

To understand the breakdown mechanism in such complex fluids, one needs to investigate also the dielectric response of magnetic fluids at electric field intensities below the breakdown value. Moreover, important information on polarization and relaxation mechanisms can be obtained by studying the dielectric response in a broad frequency scale. Thus, even though the power transformers operate at 50 or 60 Hz frequency, it is desirable to investigate the magnetic fluids dielectric properties in a broader frequency range, too. Herein we present the dielectric response of a thin layer of a transformer oil-based ferrofluid within the frequency range from 1 mHz up to 10 kHz, measured by an LCR meter.

1. Experimental. The investigated ferrofluid consists of a transformer oil (Mogul Trafo CZ-A, Paramo, CZ) and iron oxide nanoparticles with oleic acid as a surfactant. The used transformer oil constitutes a high quality liquid insulator with the DC electrical conductivity of a few fS/cm at room temperature. Its dielectric permittivity was found to be 2.1 in the whole investigated frequency range. The nanoparticles were synthesized by a well-known chemical co-precipitation method from ferrous and ferric ions, whereas the ferrofluid preparation procedure followed the well-established protocol [17]. The mean particle diameter determined by transmission electron microscopy was 10 nm. The magnetic properties of the ferrofluid were measured by a SQUID magnetometer. From a magnetic point of view, the studied magnetic nanoparticles are of superparamagnetic nature, as confirmed by the magnetization measurements (Fig. 1). At room temperature the magnetization of saturation is 29.6 $\mathrm{emu\,cm^{-3}}$ with a corresponding magnetic volume fraction of 6.6%.

The frequency dependent dielectric response of the studied system was measured in commercial liquid crystal cells. The sandwiched type cells were composed of two flat glass pieces coated with Indium Tin Oxide (ITO) conductive layers acting as electrodes. In order to minimize the errors due to edge effects at the sample-capacitor borders, the electrode separation distance in the cell was set to

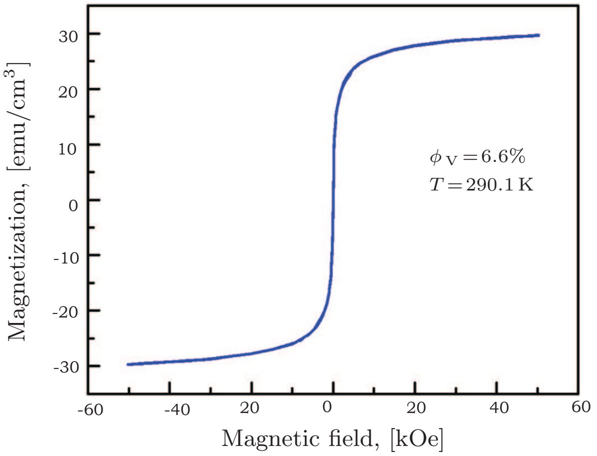

Fig. 1. Magnetization curve of the studied magnetic fluid at room temperature.

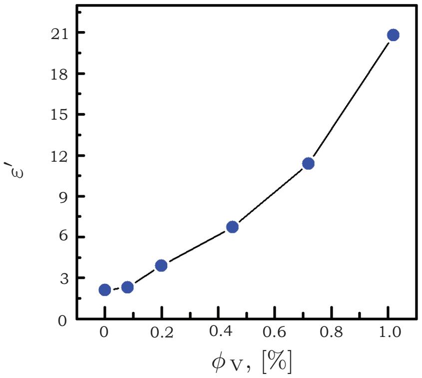

Fig. 2. Relative permittivity of the studied ferrofluid vs. the magnetic volume fraction.

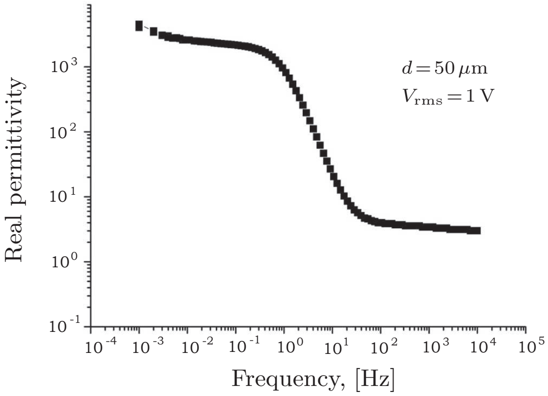

Fig. 3. Real permittivity spectrum of the investigated ferrofluid.

be $50\,\mu$m. The active electrode area was 25 (5×5) mm^2. Firstly, we measured the capacity of the empty cell and found the value of 7.5 pF. Then the cell was filled with the investigated sample by capillary forces. The transparent ITO layers allowed us to check the homogenous filling of the cell. The dielectric spectroscopy measurements were carried out by an LCR meter (IM3533 HIOKI), with an effective voltage value of 1 V across the capacitor.

2. Results. It has been proven that the relative permittivity of the transformer oil measured at a constant low frequency increases with increasing magnetic particle volume fraction in the medium. This behavior can be clearly seen from the graph in Fig. 2 which presents the relative permittivity values measured at a frequency of 20 Hz. The increasing ferrofluid permittivity reflects the increasing ability of the colloidal system to store the electrical energy. This is associated with the electrical polarizability of the iron oxide nanoparticles which is considerably higher than that of the transformer oil molecules. As it is presented in the next graphs, the dielectric dispersion appears around this frequency. It was shown [18] that this dielectric dispersion shifted towards higher frequencies with increasing particle concentration. This shift is rather nonlinear. Thus, the taken permittivity values at 20 Hz also exhibits the nonlinear dependence on the increasing particle concentration.

The dielectric spectroscopy measurements yielded the spectra of the complex permittivity of the ferrofluid. The obtained spectrum of the real permittivity is presented in Fig. 3. One can clearly see the pronounced dielectric dispersion between 10^{-1} to 10^1 Hz. Taking into account the low polarizability of the transformer oil and the fast polarizability of nanoparticles, one can assume that this dispersion is associated with a particular interfacial polarization mechanism. Since ferrofluids consist of different dielectric materials, the interfacial relaxation processes, known as the Maxwell-Wagner effect that accounts for charge accumulation at the material interfaces, is frequently taken into account when explaining such a spectrum. However, on the basis of the Maxwell-Wagner model it was estimated that the maximum of dielectric loss should appear at much higher frequencies [19] for systems like the investigated magnetic fluid. This implies that the Maxwell-Wagner theory cannot be applied to explain the obtained dielectric spectrum. To elucidate the low frequency relaxation process occurring in the studied sample, we apply the Schwarz model which was also used in several previously published works focused on the transformer oil-based magnetic fluids [18, 19]. According to this model, the low frequency relaxation in electrolyte solutions is often attributed to the polarization of counterions at the particle-solution interface. Although the transformer oil-based magnetic fluids are not ionic magnetic fluids, the co-precipitation particle preparation procedure can finally result in a colloid containing residual hydrated cations, which is practically very difficult to remove completely from the solution. Another ion impurity is present in the transformer oil in the form of moisture and additives. These ions then can act as counterions in relation to the adsorbed OH^- groups on the particle surface and create thus an electric double layer on the particles. Then the deformation of the counterion atmosphere is the main polarization mechanism in the low frequency electric field. Hence, this relaxation process causes a dielectric loss reflected in the maximum of the imaginary permittivity spectrum around 1 Hz (Fig. 4).

In Fig. 4, below the relaxation maximum, one can see a remarkable linear increase in loss factor with the decreasing frequency. This increase in loss factor is indicative of a space charge loss. Especially for thin colloidal films with the thickness in the micrometer range, the dielectric losses at very low frequencies are governed by a space-charge polarization mechanism. The considered space charges are usually assumed to be ions. However, it is well known that electrons or holes may also give rise to similar phenomena as those observed with ions. In highly purified liquids, electronic losses may predominate, but in the case of colloidal dispersion like the investigated magnetic fluid, their contribution is obscured by the very much higher ionic losses related to the presence of the free unwashed ions. On the other hand, the ions can also arise as a result of oxidation or presence of other extraneous electrolytic substances in transformer oil. As a result of the space charge migration, electric double layers are formed on the electrodes leading to the electrode blocking effect observed in the presented graph.

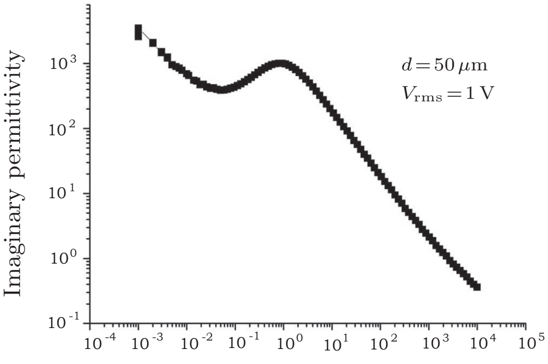

Fig. 4. Imaginary permittivity spectrum of the investigated ferrofluid.

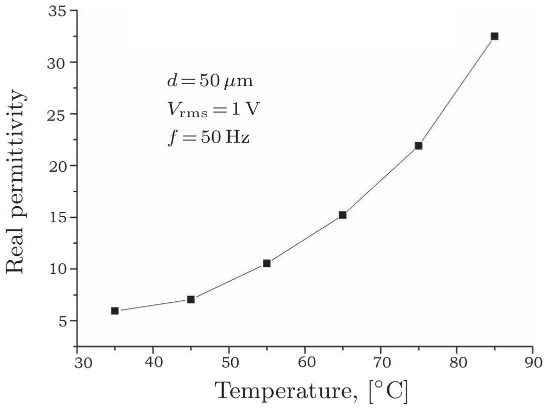

Fig. 5. Temperature dependence of the ferrofluid real permittivity measured at 50 Hz.

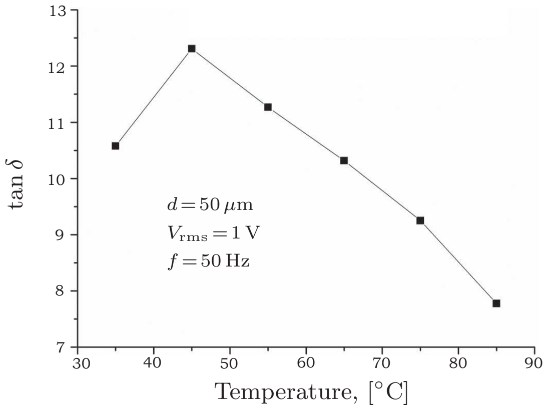

Fig. 6. Temperature dependence of the ferrofluid dissipation factor measured at 50 Hz.

It is known that the relaxation time of the observed relaxation process obeys the Arrhenius law. Therefore, with increasing temperature, the relaxation time and the related dielectric dispersion shift to higher frequencies proportionally. Then in regard to the operating frequency of the power transformers, one can find that the real permittivity of the ferrofluid increases with increasing temperature. This behavior is illustrated in Fig. 5 by plotting the selected permittivity at a frequency of 50 Hz versus the sample temperature. The increasing 50 Hz permittivity reflects the shifting trend of the real permittivity plateau and the related dispersion in the measured dielectric spectrum. In fact, this is associated with the increasing mobility of the counterions due to the increasing thermal energy. On the other hand, the relaxation maximum shifts to higher frequencies with increasing temperature, too. However, as its position at room temperature is around 50 Hz, it is clear that the further shift to the higher frequencies will result in a reduced loss factor as well as in a dissipation factor ($\tan\delta = \varepsilon''/\varepsilon'$) which is determined as the ratio of the imaginary and real permittivity values. The temperature dependent dissipation factor is depicted in Fig. 6.

3. Conclusion. We have studied the dielectric response of a transformer oil-based magnetic fluid in the broad frequency range. One interfacial relaxation process was found in the low frequency region, whereas the electrode polarization effect was observed at lower frequencies. Both phenomena are associated with the presence of ionic impurities in the oil and at the particle-oil interface. The presented high values of the real permittivity can suggest an application potential of such colloidal systems in the field of super capacitor technology. However, high dielectric losses may constitute a real drawback when trying to apply the studied magnetic fluid in power transformers. It was shown that the dielectric permittivity and the dissipation factor of the studied magnetic fluid measured at the transformer operating frequency are remarkably dependent on the sample temperature.

Acknowledgements.
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