Dielectric Response at Different Nanoparticle Concentrations for GTL Oil-based Magnetic Nanofluids

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Abstract: Mineral oil, a petroleum product, has been utilized as transformer oil for over a century. The advent of novel technologies in gas processing, has engendered novel opportunities in liquid insulations. Gas-to-liquid (GTL) processing offers a stable insulating liquid suitable for use in transformers. The present work focuses on studying of the dielectric properties of magnetic nanofluids where GTL oil is the carrier fluid. Samples of nanofluids with different concentrations of iron oxide nanoparticles have been produced. Frequency- and time-dependent spectroscopic methods were utilized as observational tools. The polarization and depolarization current (PDC) method was used to study the dielectric properties when applying a direct current (DC) electric field with an intensity of 0.1 kV/mm and 1 kV/mm, respectively. The results obtained from this study demonstrated an apparent enhancement in conduction current values and a temporal variation in absorption current with an increase in nanoparticle concentration. In the case of frequency-dependent spectroscopy, changes in relaxation processes were observed and changes in material polarization due to increased nanoparticle concentration were determined. The dielectric method employed the real and imaginary parts of the dielectric modulus, to express the polarization behavior.

Keywords: nanofluid; nanoparticle; dielectric spectroscopy; insulating material; transformer

1 Introduction

The increasing electrification of industry and households has resulted in greater demand for electricity transmission and distribution equipment. Among this equipment, transformers are of particular technical and economic value [1]. These devices are utilized at both the transmission and distribution levels, are engineered for long service, and are difficult to replace in the event of failure [2]. The present study has revealed that most transformers have a combined mineral oil-paper insulation system. Since mineral oil is a petroleum product, it must be replaced by alternative technology that exhibits long life, good insulating and thermal properties, and, moreover, must be biodegradable. In addition to using biodegradable transformer oils, a future possibility is the implementation of nanoparticles in insulating fluids [3] [4]. This solution offers an increase in equipment cooling efficiency as well as an increase in dielectric properties. For the last two decades, research in this field has been progressive, with researchers and engineers focusing on developing stable insulating fluids enriched with the contribution of nanoparticles.

1.1 Transformer Oil-based Magnetic Nanofluids

Transformer oils used in the electrical power industry are commonly based on mineral, synthetic, or natural ester bases. These oils have excellent electro-insulating properties and can efficiently dissipate waste heat generated in electrical power devices such as transformers, reactors, and similar equipment. However, mineral oils can degrade at high temperatures, reducing their performance and lifespan. Therefore, natural esters are increasingly used as they are biodegradable, non-toxic, and have better oxidation resistance. The oil degradation poses a risk to the oil and the insulation system of oil-paper or other composite components of the electrical device insulation systems.

Aside from the nature of the insulating oil, as mentioned, the most crucial property is its ability to dissipate waste heat by conduction or convection [5]. This property and enhancing dielectric strength can be improved by creating nanofluids based on these oils [6]. One of the most commonly used methods for producing insulating oils is the GTL (Gas-to-Liquid) method, which allows the production of transformer oil from natural gas. This process has several advantages, such as better purity and improved insulating properties than traditional mineral oils. In the context of nanotechnology, the GTL preparation of transformer oil focuses on enhancing its basic properties, such as dielectric strength, thermal conductivity, and resistance to degradation at high temperatures [6]. This technology provides a foundation for effectively integrating nanoparticles into the oil, creating of nanofluids with enhanced performance characteristics.

Nanofluids created this way are liquids in which nanoparticles, ranging in size from 1 to 100 nanometers, are dispersed. In the case of transformer oil, these nanoparticles are typically metallic or oxidative materials such as Fe_3O_4 (iron oxide), TiO_2 (titanium dioxide), or Al_2O_3 (aluminum oxide), which improve the insulating and thermal properties of the oil [7]. The preparation of these nanofluids involves these key steps:

Dispersion of Nanoparticles into the Base Oil

Nanoparticles are initially dispersed in the liquid medium using various techniques such as ultrasonic dispersion, magnetic mixing, or high-pressure homogenization.

Control of Particle Concentration

The concentration of nanoparticles in the oil affects its insulating properties and thermal and mechanical behavior. The optimal concentration depends on the type of particles and the system's performance requirements. A suitably chosen concentration of nanoparticles in the carrier fluid can influence the electrical and thermal properties of insulating fluids. In the case of iron oxide nanoparticles, the electrical strength can be improved by a factor of 1.5 to 2 in samples with a surfactant. It is the surfactant that plays a role in preventing sedimentation and agglomeration of nanoparticles [8]. When using nanoliquids in transformers, it is necessary to follow the existing rules for sufficient distances between high-voltage and grounded parts. Any agglomerates in the form of chains cannot bridge these distances (dimensions in tens of um) due to the action of an alternating electric field. In addition, there is natural circulation of liquid in the transformer, which creates pressure on such chain agglomerates or can carry them away [9].

Nanofluids can reduce the operating temperature of devices due to the phenomenon of thermomagnetic convection, where the combined effect of the temperature gradient and magnetic field creates a complex process in which temperature and the magnetic field affect fluid flow. This phenomenon can cause the formation of convection currents that form in different ways depending on the intensity of the magnetic field and temperature gradient [10] [11].

1.2 Preparation of Nanoparticles in General

Nanoparticles are produced using various methods, with the most common being:

Co-precipitation: This method is very popular for producing magnetic nanoparticles like Fe_3O_4. It allows for the creation of particles with high stability and controlled size [12].

Sol-gel process: This method is effective for producing ceramic or metal oxide nanoparticles, which can improve a nanofluids' mechanical and thermal properties [12].

Ultrasonic dispersion: After the nanoparticles are produced, it is essential to disperse them in the liquid to achieve maximum stability evenly. Ultrasonic dispersion helps create a uniform suspension without forming of agglomerates, which could reduce the fluid’s effectiveness [12].

An important aspect of nanoparticle preparation is surface modification, which ensures their stability in the liquid. Without surface modification using surfactants, particles would quickly sediment or agglomerate, degrading the colloidal stability of the liquid and thus negatively affecting the insulating and mechanical properties of the nanofluids [13].

1.3 Role of Surfactants

Surfactants play a key role in stabilizing nanofluids. These substances reduce the surface tension between nanoparticles and the liquid, preventing sedimentation or agglomeration of particles. Using surfactants like oleic acid or various types of nonionic or anionic surfactants enhances the stability of the liquids at higher temperatures and in aggressive environments. These substances improve the dispersion of nanoparticles, achieving better homogeneity of the nanofluid and its desired properties.

Additionally, surfactants can improve the liquid's thermal and mechanical properties because stabilized particle dispersion allows for better heat transfer and prevents the formation of microcracks in the oil at high voltages [14].

1.4 Dielectric Spectroscopy Techniques

Dielectric spectroscopy can be defined as the measurement of a material's fundamental dielectric properties as a function of time or frequency. In the research of dielectric materials, such as insulating oils, frequency-dependent dielectric spectroscopy (FDS) and time-dependent dielectric spectroscopy (TDS) methods are frequently applied [15-17].

The primary advantage of these methods is that they are non-destructive, minimally instrumentation-dependent, and can be executed relatively quickly. Among the spectrum of TDS methods, the polarization and depolarization current (PDC) method is among the most popular. This method involves applying of a direct current (DC) voltage to the sample, with the measuring apparatus monitoring the evolution of the current flowing through the sample. Subsequently, the voltage source is switched off and the apparatus (ammeter) monitors the discharge current waveform. The resulting waveforms offer insight into the conduction and polarization loss contributions, which are reflected by the nature of the behaviors. However, this method is hindered by small currents, which are susceptible to interference from the surrounding environment [18] [19].

The limitations of the PDC method can be addressed by employing the FDS method. This method does not require such a high measurement voltage amplitude, is more resistant to ambient interference and takes a relatively short time (except at low frequencies). The FDS principle is founded on implementing a sinusoidal measurement voltage of varying frequencies, at which the amplitude and phase shift of the current are measured [20]. After this, the geometry of the electrode system can be utilized to calculate parameters such as sample capacitance, loss factor, and components of the complex permittivity[21]. To achieve optimal characterization, the TDS and FDS methods can be supplemented by temperature control, given the temperature-dependent nature of the dielectric properties of the materials. These methods facilitate analyzing relaxation processes, electrical conductivity, and material sensitivity to degradation factors [22-24].

2 Materials and Methods

2.1 Preparation Magnetic Nanofluids

The insulating liquid Shell Diala S4 ZX-I, a GTL (gas-to-liquid) technology product, was utilized as the carrier oil to synthesize of nanofluids. The S4 ZX-I exhibits a sulfur content of less than 1 ppm, as defined by ASTM D5185 [25]. The examined samples contain superparamagnetic iron oxide (predominantly Fe_3O_4) magnetic nanoparticles. The magnetic nanofluid procedure has been documented in references [26] [27]. This type of nanofluid can't contain a single type of iron oxide. Magnetite (Fe_3O_4) slowly changes into γ-Fe_2O_3. This process is explained in more detail in [28]. For optimal characterization, the implementation of iron oxide is generally preferred. Oleic acid is employed as a surfactant for the nanoparticles within the carrier liquid, coating their surfaces and hindering the formation of clusters. The mixing ratio of the carrier liquid and the raw nanofluid was systematically varied to generate samples with varying nanoparticle concentrations. The raw magnetic nanofluid with the manifested magnetic properties is presented in Figure 1.

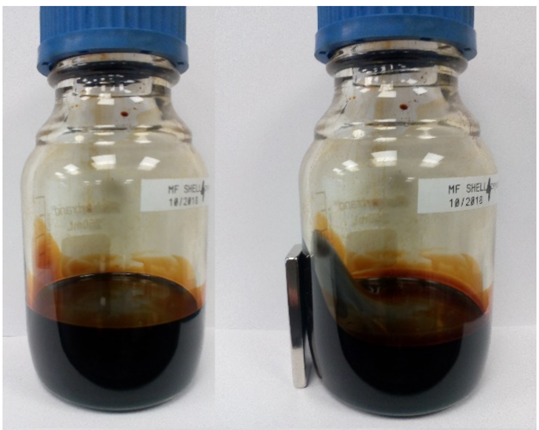

Figure 1
Raw magnetic nanofluid is prepared by mixing samples of different concentrations. An NdFeB magnet demonstrates nanofluid magnetic properties.

The nanoparticle size distribution was verified using a method reported in a similar experiment for hybrid nanofluids [29]. The mean hydrodynamic diameter of the nanoparticles was determined to be 13.3 nm by the dynamic light scattering method in [30]. The determination of a polydispersity index value of 0.21 was calculated. Five samples with bulk nanoparticle concentrations were created, in Table 1.

Table 1
Concentrations of iron oxide NPs in investigated samples

Sample	**S0**	**S1**	**S2**	**S3**	**S4**	**S5**
Vol. concetrations MNPs [%w/V]	base oil S4 ZX-I 0.00	0.05	0.1	0.5	1.0	2.0

2.2 Dielectric Measurements

Dielectric measurements of nanofluids were performed using dielectric spectroscopy methods in the frequency domain and time domains. The individual measurements were carried out under laboratory conditions with an air temperature of 295 K, an air pressure of 1017 hPa, and a humidity of 32 vol%. The samples were placed in a Keysight 16452A liquid dielectric test fixture, containing two electrodes (Fe 54%, Co 17%, Ni 29%) separated from the conductive parts an Al_2O_3 insulator. The spacing of the interelectrode space was defined by a 1mm spacer and the total sample volume was 4.8 ml. In this case, the electrode housing functions as the third grounding electrode. Using a 3-electrode system enables the acquisition of verifiable results by suppressing parasitic capacitances [31]. The order of the experiments was FDS first, followed by TDS, to minimize the influence of polarization on the sample, as previously described in [17]. In the case of TDS, the polarization/depolarization currents (PDC) method was used. The test fixture was connected to the measurement systems using a 16048A test lead (BNC-to-BNC). The experimental setup with the electrode system for FDS and PDC methods is shown in Figure 2.

2.2.1 Frequency Domain Spectroscopy

FDS was performed using an IDAX 300 (Megger, Sweden) with an applied alternating test voltage 2V RMS. The frequency range of the IDAX 300 extends from 0.1 mHz to 1 kHz. Each sample was measured thrice, and the mean of these measurements was used to identify outliers. After each measurement, the measuring apparatus was short-circuited and connected to a ground terminal in the laboratory. After 30 minutes, an additional measurement was taken.

2.2.2 Time Domain Spectroscopy

In the case of TDS, the polarization/depolarization current (PDC) method was used. The internal source of the electrometer 6517B (Keithley Instruments Inc., USA) was used as the electric field source. The sample was subjected to a homogeneous DC electric field with an intensity of 0.1 kV/mm and 1 kV/mm for 1000 s. Subsequently, the electric field source was disconnected, and the depolarizing current was measured at a further 1000 s. Current values were recorded using a Keithley 6517B integrated ammeter, with a sampling rate of 1 sample per 100 ms. It should be noted that only five samples were taken for each second [32]. Three measurements were taken for the PDC for each nanofluid sample, and the resulting value was the arithmetic mean. After each measurement, the apparatus was short-circuited and connected to the laboratory's ground terminal to eliminate space charge.

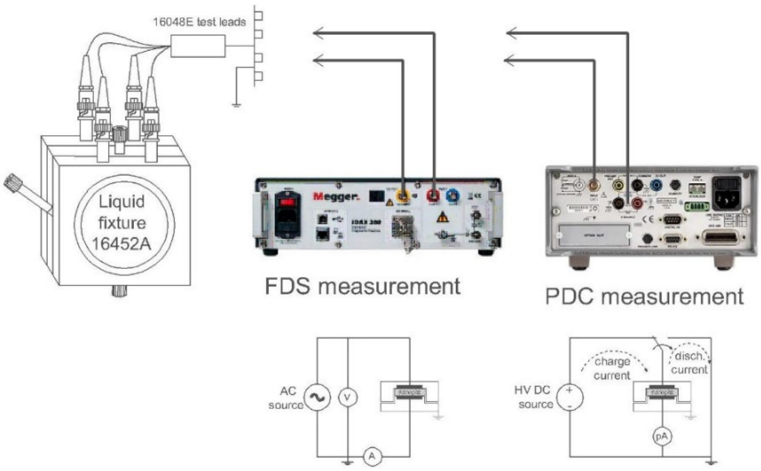

Figure 2
Test setup for FDS and PDC test methods with schematic diagrams

3 Results and Discussion

3.1 Dielectric Frequency Response of GTL Oil and Magnetic Nanofluid based on GTL

The dielectric response of pure GTL-based transformer oil in the form of complex permittivity is presented in Figure 3. It is evident from the data presented that the real component in the case of pure oil exhibits frequency independence.

The measured data are close to 2, a typical value for transformer oils. In the case of nanofluid with a concentration of 0.05%w/V, frequency independence occurs only in the region from 0.1 Hz. The observed decrease in the real part of the complex permittivity indicates the presence of a relaxation mechanism within the nanofluid sample, operating within the range of 0.1 mHz to 0.1 Hz. This region is designated as the dielectric dispersion region. In the absence of a dielectric dispersion region in the case of pure carrier oil, this aspect will not be further explored in the subsequent interpretation of the results.

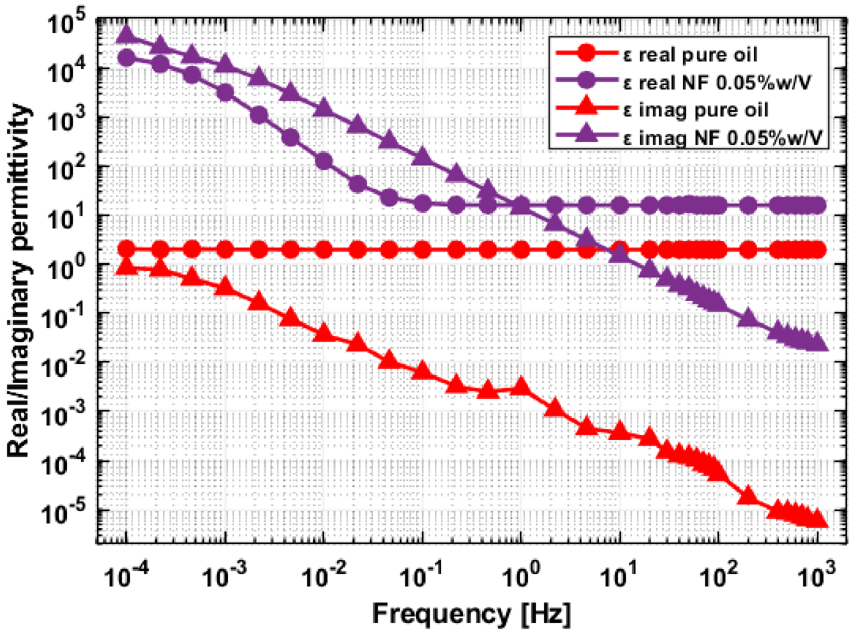

Figure 3

Real and imaginary permittivity of pure GTL oil and magnetic nanofluid

The nanofluid's imaginary part of the permittivity does not typically display the characteristic Debye behavior. This phenomenon may be attributed to the superposition of the significant contribution of the sample conductivity and electrode polarization. In the case of magnetic nanofluid, the formation of nanoparticle around the electrodes is a key observation. This layer exhibits an additional capacitance in the low-frequency regions, reducing the current that passes through the nanofluid sample. The frequency and temperature dependence of electrode polarization in the case of nanofluids is suitably described and investigated in [21].

3.2 Effect of Different Concentrations Nanoparticles on Dielectric Frequency Response of GTL Oil-based Magnetic Nanofluid

From the above facts, in the FDS method results, the influence of electrode polarization should be eliminated for better interpretability of the results. A suitable tool is the complex dielectric modulus. Following the findings of specific studies [33], it has been determined that the complex dielectric modulus constitutes a suitable interpretation tool for the analysis in the context of FDS, given its ability to effectively suppress the influence of electrode polarization in the characteristics of complex permittivity elements, thus enabling the accurate and precise determination of those mentioned above complex dielectric modulus. This is defined as the inverse of the complex permittivity and can be expressed by equation:

$$M^*(\omega) = \frac{1}{\varepsilon^*} = \frac{\varepsilon`(\omega)}{\varepsilon^{r2}(\omega)+\varepsilon^{rr2}(\omega)} + \frac{i\varepsilon`(\omega)}{\varepsilon^{r2}(\omega)+\varepsilon^{rr2}(\omega)} = M`(\omega) + iM``(\omega) \quad (1)$$

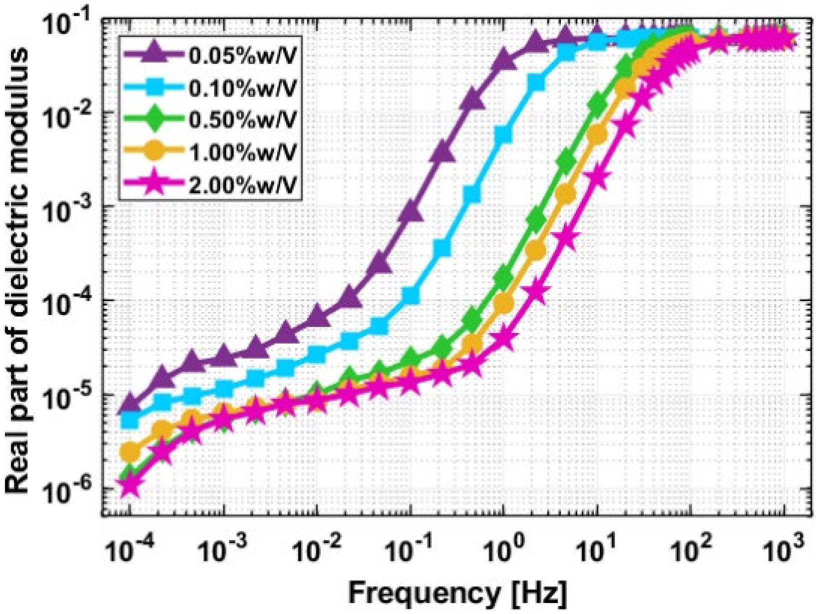

Figure 4
The frequency spectrum of the real part of the dielectric modulus for different nanoparticle concentrations in the NF

The dielectric modulus of magnetic NFs with different concentrations is presented in Figures 4 and 5. In Figure 4, a general decrease in the values of the real component of the complex dielectric modulus with increasing nanoparticle concentration in the sample is evident. In addition, the dielectric dispersion region shifts towards higher frequencies due to increased nanoparticle concentration.

Within the higher frequency range (100 Hz – 1 kHz), a uniform level of the real part of the modulus is evident across all samples. This phenomenon can be attributed to the utilization of GTL oil as the carrier fluid in all samples. In the case of the real part of the complex permittivity, the values in this region are close to those of pure GTL oil. Still, they are increased by additional compounds such as surfactant or the permittivity of the nanoparticles themselves.

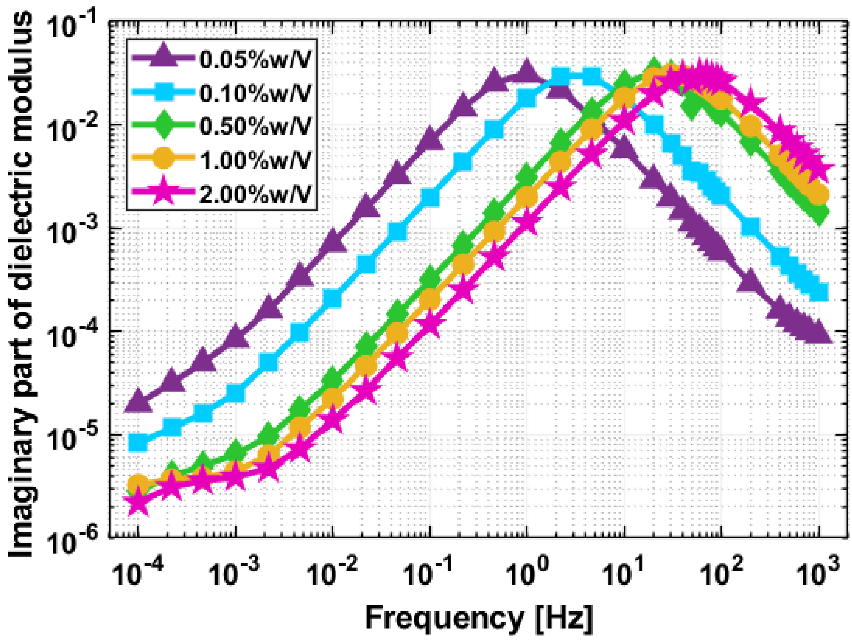

Figure 5

The frequency spectrum of the imaginary part of the dielectric modulus for different nanoparticle concentrations in the NF

In the case of the imaginary component of the dielectric modulus in Figure 5, a shift of the characteristics towards the higher frequency region is also evident. The contribution of the nanoparticles, their surfactant, and the impurities in the samples due to the nanoparticle manufacturing process cause the samples to exhibit a polar character. After applying an alternating electric field to the sample, it becomes impossible for polarization events to follow rapid changes in the electric field immediately. This delay is attributed to relaxation, characterized by the accumulation of charges on the nanoparticle surface. In the case of nanoparticles dispersed in a liquid, trapping of free charge on the nanoparticle's surface is possible. This forms an electrical double layer on the nanoparticle surface, thereby conferring a polar character on the nanoparticle. The ensuing relaxation process manifests characteristics consistent with the Cole-Cole relaxation model. Gaussian-like curves represent the relaxation process in the results presented here. The peaks of the individual behaviors shift to the right, to regions of higher frequencies, as samples with higher nanoparticle content are more polar. According to [34], nanoparticles can bind water molecules to themselves, which may have entered the sample from atmospheric moisture. It is hypothesized that, in such instances, the higher the concentration of nanoparticles, the greater the capacity for water molecule binding, with these water molecules being absorbed by the nanoparticles' surface. However, as other studies show, the influence of the double-layer effect on the surface of nanoparticles is much more pronounced [35] [36].

The determination of the frequencies of the individual peaks of the relaxation events was made possible by the waveforms of the imaginary part of the dielectric modulus, as illustrated in Figure 5. After this, the f_{peak} frequency values were plotted on the graph presented in Figure 6. This figure demonstrates the dependence of the peak frequencies on the nanoparticle concentration. A simple fitting line for a 1st-order polynomial has been constructed. A robust least-squares fitting method with square weights was used. The coefficient of determination for this type of fitting was R2 = 0.9955. The resulting equation is written as:

$$f(x) = f_{peak} = 30.3617 \cdot Concentration - 0.6091 \quad (2)$$

The resulting regression shows a strictly linear dependence of the relaxation process maxima on the nanoparticle only in the study range of concentrations in the nanofluid.

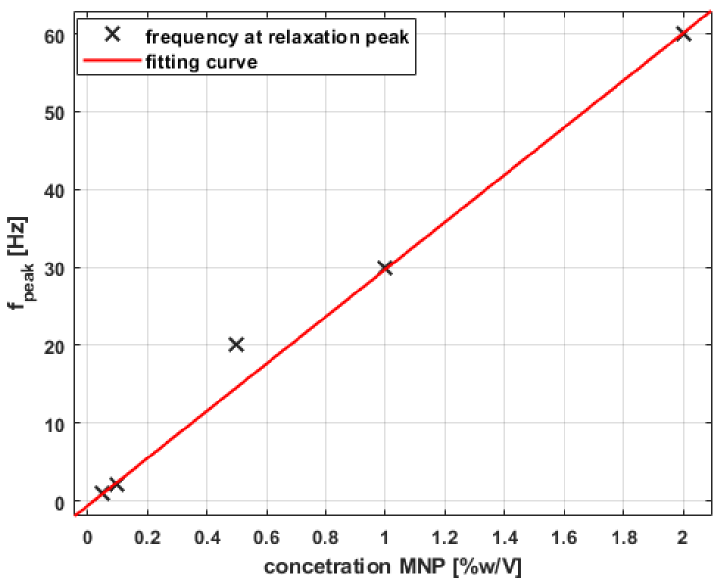

Figure 6

Fitting curve of the frequency domain characteristic peak frequency and concentration of iron oxide nanoparticles

3.3 The Effect of Varying Concentrations Nanoparticles in GTL Oil-based Magnetic Nanofluid on Behaviors of PDCs

The results of the PDC experiments on pure GTL oil and GTL-based magnetic nanofluids subjected to different DC electric field strengths are shown in Figures 7 and 8. Figure 7a shows the polarization currents flowing through the samples at an electric field strength of 0.1 kV/mm. At first sight, a significant difference between the pure GTL oil sample and the nanoparticle-modified samples can be seen. However, when nanoparticle concentrations in the nanofluids were increased tenfold (0.05%w/V and 0.50%w/V) or tenfold (0.10%w/V and 1.00%w/V), a similar tenfold difference was observed in the charging current responses.

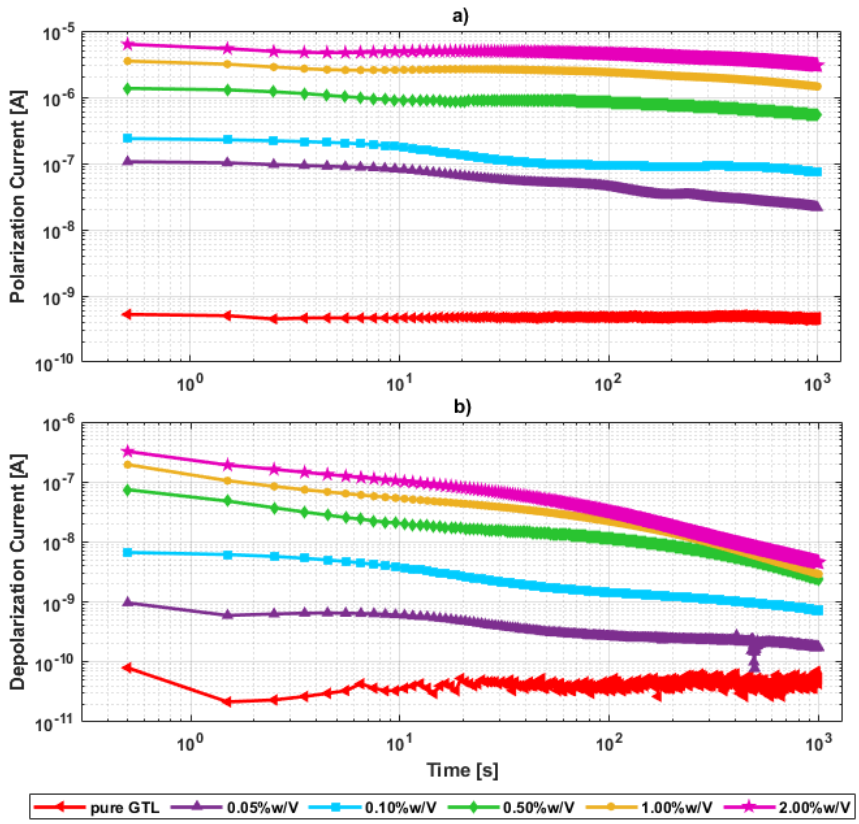

Figure 7
PDC behaviors with applied electric intensity 0.1kV/mm. Dependence of polarization current a) and depolarization current b) on time

As illustrated in Figure 7b, the depolarization current waveforms demonstrate a substantial dispersion of values for the pure GTL oil sample. This variability might be attributable to the proximity to the upper limit of the measuring apparatus (approx. 40 pA for pure GTL). The most significant declines are evident for the 1.00%w/V and 2.00%w/V concentrations. This phenomenon can be attributed to the gradual release of trapped free charges on the nanoparticle surface. The hypothesis can be further substantiated by the above FDS results, which demonstrate that a higher nanoparticle concentration in the sample is associated with a double electric layer on the nanoparticle surface and the ability of nanoparticles to bind water molecules.

A higher value of charging currents was exhibited after applying an electric field of 1 kV/mm to the liquid samples. The waveforms of polarization currents are displayed in Figure 8a, where the difference between nanofluids and pure GTL oil can be observed. However, this difference was less significant when an electric field of 0.1 kV/mm was applied.

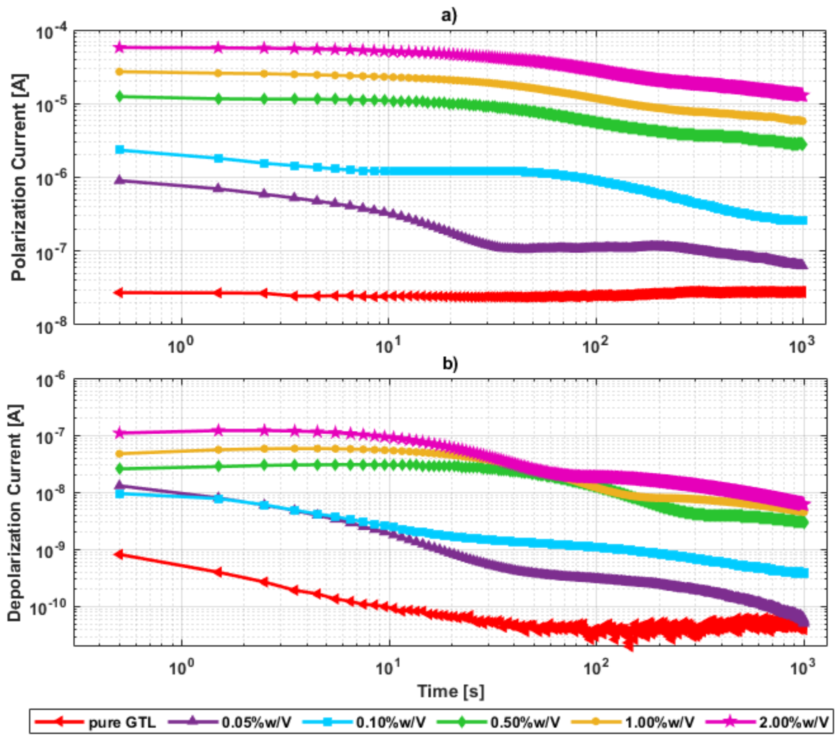

Figure 8

PDC behaviors with applied electric intensity 1.0 kV/mm. Dependence of polarization current a) and depolarization current b) on time

A slight decrease can be observed in the region of 10-100 s region when a stronger electric field is applied to the nanofluid sample. This decrease can be attributed to free charge carriers trapping on the surface of the nanoparticles and electrodes, which dynamically changes the capacitance ratios in the sample. Notably, a more significant value of the electric intensity will accentuate these dynamic processes, which also take place in samples loaded with a weaker electric field, but are less pronounced in the waveforms. Another hypothesis dielectrophoretic action, which is investigated at high DC voltage values in [37]. Although the authors demonstrate aggregation of nanoparticles in a nanofluid with a concentration of 0.5%, they do not observe this at voltages less than 9 kV. However, lower voltages (0.5-2 kV) have been considered in some of the simulations.

In the presented work, the polarization current characteristics show high values of the conduction component of the current for the GTL oil modified by iron oxide nanoparticles. The following DC conductivity has been determined according to the known relationship:

$$\sigma_{DC} = \frac{\varepsilon_0}{C_0 U_0}\left(i_{pol}(t) - i_{depol}\right); [pS \cdot m^{-1}] \tag{3}$$

Where C_0 is the geometrical capacitance, ε_0 is the free-space permittivity, and U_0 is the applied voltage to the sample. The resulting values are then plotted against each other to create Figure 9, which shows the dependence of the DC conductivity on the change in nanoparticle concentration in the magnetic nanofluids.

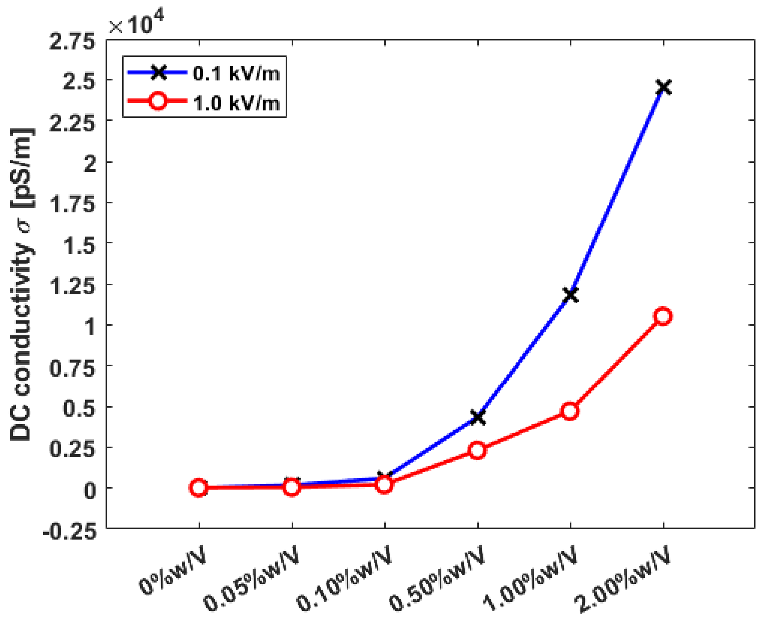

Figure 9
Dependence of DC conductivity on the concentration of nanoparticles in GTL-based nanofluid

From other studies, it can be assumed that the conductivity of the sample will increase with the nanoparticle content [38]. In the case of Fe_3O_4 nanoparticles, according to [39], a mechanism of conductivity at low frequencies is described, where electrical conductivity is associated with long-range mobility and grains with high resistance. In [40] the variation of electrical conductivity of nanofluids as a function of nanoparticle concentration is studied. The work investigates water-based nanofluids, where the conductivity is higher than that of GTL-base fluid by a factor of several times (3 µS vs. 30 pS) but the trend of conductivity increase due to nanoparticles is the same.

Conclusions

The measurement results demonstrate that the presence of iron oxide nanoparticles in GTL transformer oil has a notable effect on the conductivity of the resulting nanofluid. This phenomenon is most evident in time-dependent measurements. The frequency-dependent method can be used to observe the polarizability of nanofluids as a function of nanoparticle concentration. The observed manifestations of the samples, with increasing nanoparticle concentration, can be explained by electron trapping mechanisms and the presence of free charge carriers, which are naturally associated with by-products, from the nanofluid and nanoparticle synthesis process.

The results motivate further investigation of the polarization and conduction processes in nanofluids with lower concentrations. As indicated in the work, higher nanoparticle concentrations create conditions that are undesirable from the point of view of transformer operators. However, the unique properties of these colloidal fluids, have stirred significant research in this area.

Acknowledgement

This work was supported by the Slovak Academy of Sciences and Ministry of Education in the framework of projects VEGA 1/0380/24 and VEGA-2-0029-24 and the Slovak Research and Development Agency under contract No. APVV- 22-0115.

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