

Temperature dependences of parameters characterizing dielectric properties of transformer oil with magnetic liquid impurities

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Abstract. The dielectric properties of pure transformer oil and transformer oil with a magnetic liquid impurity with a concentration of 4.2 vol.% have been investigated in the frequency range of 6 to $7 \cdot 10^5$ Hz and at the temperatures of 308 to 423 K. The dielectric permittivity of the transformer oil has been shown to increase almost linearly with temperature. The dielectric permittivity of the transformer oil with the magnetic liquid impurity has exceeded the dielectric permittivity of the transformer oil without impurities and depended nonlinearly on temperature. The electrical conductivity of the transformer oil without impurities in the entire frequency range and the transformer oil with the magnetic liquid impurity at the frequencies $f > 3 \cdot 10^3$ Hz at a specific temperature have been shown not to depend on the frequency. These values of the electrical conductivity reflect the bulk properties of the liquid. The temperature dependence of the electrical conductivity has been shown to correspond to the Arrhenius law. It has been found out that the activation energy of the temperature dependence of the electrical conductivity of the pure transformer oil is 0.27 eV, and 0.22 eV at presence of the magnetic impurity. The dispersion of the values of the real and imaginary components of the complex dielectric constant of the transformer oil with the magnetic liquid impurity corresponds to the Cole–Cole dispersion. Based on the analysis of the Cole–Cole diagrams, the values of the relaxation time τ , the thickness of the near-electrode region W , and the parameter characterizing dispersion of the relaxation times α have been found for each of the sample temperatures. The value of τ has been shown to decrease with temperature, the value of W increases at increasing the temperature, and the value of α equals as 0.06 and has no temperature dependence.

Keywords: dielectric permittivity, electrical conductivity, relaxation time, transformer oil, magnetic liquid impurities.

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1. Introduction

At present (and apparently in the near future), electrical energy will be the main energy type used by people. This is due to the fact that the electrical energy can be relatively easily transformed into other energy types.

When consuming the electrical energy, the optimal situation would be if each consumer had a source of electricity. This is especially true for generators based on conversion of energy received from the Sun into the electrical energy. However, such sources are still very expensive, although their cost constantly decreases.

When using various types of powerful power plants, a rather complex problem of transmitting electricity over a certain distance arises, since power plants (especially nuclear ones) must be built at a considerable distance from electricity consumers. At present, effective transmission of electricity over a certain distance is realized by increasing or decreasing voltage with transformers. Since transformers heat up during operation, one of the important technical and scientific issues is ensuring their effective cooling.

More efficient cooling of transformer windings can be achieved by introducing magnetic liquid into the transformer oil (as a transformer winding cooler). When using only the transformer oil, cooling occurs mainly due to heat transfer by convection flows in the liquid. In the presence of a magnetic impurity, the speed of movement of the liquid (and, hence, the increase in the cooling efficiency) can be significantly enhanced due to the action of a magnetic field on the magnetic liquid particles. To significantly increase the efficiency of the transformer cooling in the presence of a magnetic liquid, it is important to establish how such an impurity influences the dielectric properties of the transformer oil.

Analysis of publications has shown that a fairly large number of studies have been devoted to the properties of transformer oil and influence of various types of impurities on it [1–6]. However, the effect of magnetic impurities on the dielectric properties of the transformer oil has not been established.

In [7], we showed that magnetic impurities insignificantly increase the dielectric permittivity of transformer oil. However, the electrical conductivity of the transformer oil increases significantly when magnetic impurities are introduced (by 2–4 times depending on the type of impurity). It was analyzed as well how the magnetic impurity affects other parameters of the transformer oil. However, all the studies were carried out at room temperature. During operation of a transformer, the windings are heated by electric current. Therefore, an important scientific and technical task is to study influence of magnetic liquid on the dielectric properties of transformer oil at different temperatures.

Therefore, the aim of this work was to study temperature dependence of parameters characterizing dielectric properties of a transformer oil with magnetic liquid impurities.

2. Materials and methods

For the research, we used transformer oil from Shell (Shell oil). Both the transformer oil without impurities and with a magnetic liquid impurity (MF/Shell DIALA S4 Fe_3O_4 - OA) were studied. The carrier liquid used in the present study is a readily biodegradable electrical insulating oil Shell Diala S4 ZX-I manufactured from zero Sulphur oils produced using Shell's GTL (gas-to-liquid) technology. The basic physical properties provided by the manufacturer are listed in Table 1. Herein, the chosen insulating oil is labeled as "oil".

Table 1. Selected physical characteristics of the base insulating oil provided by the manufacturer.

Properties	Typical values
Density at 20 °C (kg/m^3)	805
Kinematic viscosity at 40 °C (mm^2/s)	9.8
Kinematic viscosity at –30 °C (mm^2/s)	438
Pour point (°C)	–45
Flash point (°C)	191
Interfacial tension at 25 °C (mNm)	51
Breakdown voltage IEC 60156 (kV)	50...60
Dielectric dissipation factor at 90 °C, IEC 60247	0.001

The transformer oil has been nano-functionalized by iron oxide magnetic nanoparticles (MNP). The MNP were synthesized by chemical co-precipitation method using iron (II) sulfate heptahydrate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$), iron (III) chloride hexahydrate ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$) and an ammonium hydroxide solution (NH_4OH). Oleic acid ($\text{C}_{17}\text{H}_{33}\text{COOH}$) was used as a surfactant to stabilize the MNP. All the raw materials were of analytical reagent grade without any additional purification. The preparation of the basic ferrofluid consisted of two steps. First, the MNP were obtained by chemical co-precipitation of the ferrous and ferric salts in the molar ratio 1:2 in an alkali medium at the temperature of 353 K. Then, MNP were sterically stabilized and coated by a single layer of oleic acid at the temperatures in the range of 353 to 355 K. The hydrophobic coated MNP were dispersed in the transformer oil. A stable suspension without any phase separation and sedimentation was obtained. The basic ferrofluid was finally diluted by the transformer oil to achieve the volume magnetic fraction of 4.2%.

To measure the dielectric properties, we used sandwich cells with a distance between the electrodes of 20 μm [7]. As in the case of liquid crystal studies, the measuring cell was filled with the test liquid by using the capillary method due to the small thickness of the cell. Before filling the cell with the test liquid, the capacitance of the cell was measured.

Similar to [7], we used the oscilloscope method for dielectric measurements [8]. The measurements were carried out in the frequency range of 6 to $7 \cdot 10^5$ Hz and at the temperatures of 308...323 K. The amplitude of the sinusoidal measuring signal was 5 V. Based on the analysis of each of the oscillograms, we determined the values of the parameters for a specific frequency. From these values, the resistance R and capacitance C of the sample were found. The components of the complex dielectric permittivity ϵ'' and ϵ' were determined using the known values of R and C as well as the geometric parameters of the samples.

The temperature was maintained with an error of no more than 0.2 K by using a thermostat specially designed for the intended measurements. The temperature range of 308 to 323 K, at which the research was carried out,

corresponds on average to the temperature range at which industrial transformers operate in power transmission lines. This is to some extent quite important for practical use of the results of this work.

3. Results and discussion

Fig. 1 shows frequency dependences of the real part of the complex permittivity ε' for the transformer oil (curve 1) and transformer oil with the magnetic liquid impurity (curve 2) at a temperature of 308 K. Similar dependences were also observed at other temperatures, hence, they are not shown anywhere in the paper.

As follows from the analysis of the obtained data, the value of ε' does not depend on f in the entire frequency range (from 6 to $7 \cdot 10^5$ Hz) (curve 1 in Fig. 1), which is typical for impurities-free liquids. However, for the transformer oil with the magnetic liquid impurity (curve 2 in Fig. 1), the value of ε' depends on the frequency at $f < 3 \cdot 10^3$ Hz. The reasons for this dispersion of $\varepsilon'(f)$ will be analyzed following the analysis of the frequency dependence of ε'' .

Fig. 2 shows frequency dependences of the imaginary part of the complex permittivity ε'' for the transformer oil (curve 1) and transformer oil with the magnetic liquid impurity at the temperature of 308 K. Based on the analysis of the obtained data, the following conclusions can be drawn. First, the value of ε'' for the transformer oil decreases inversely proportionally to the frequency in the entire frequency range (curve 1 in Fig. 2). The same dependence is observed for the transformer oil with the magnetic liquid impurity for the measuring signal frequencies $f > 3 \cdot 10^3$ Hz.

The inverse proportionality of ε'' to f means that the value of conductivity does not depend on frequency.

The same dependences $\varepsilon''(f)$ were observed at temperatures different from 308 K. The specific electrical conductivity is determined from the following relation:

$$\sigma_{AC} = 2\pi f \varepsilon_0 \varepsilon'', \quad (1)$$

where σ_{AC} is the specific electrical conductivity and ε_0 is the dielectric constant, respectively.

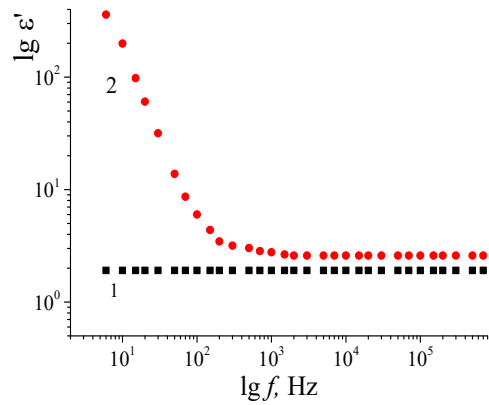


Fig. 1. Frequency dependences of the real part of the complex permittivity ε' for transformer oil (curve 1) and transformer oil with magnetic liquid impurity (curve 2) at the temperature of 308 K.

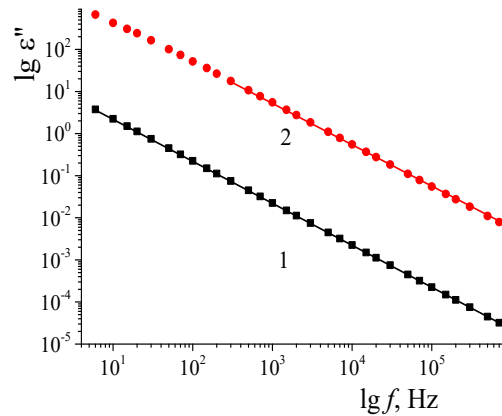


Fig. 2. Frequency dependences of the imaginary part of the complex permittivity ε'' for transformer oil (curve 1) and transformer oil with magnetic liquid impurity (curve 2) at the temperature of 308 K.

The values of the electrical conductivity σ_{AC} of the transformer oil at different sample temperatures are given in Table 2.

Table 2. Temperature dependence of electrical conductivity at alternating current σ_{AC} for transformer oil.

Temperature, K	308	313	318	323
σ_{AC} , $\text{Ohm}^{-1}\text{m}^{-1}$	$1.26 \cdot 10^{-9}$	$1.5 \cdot 10^{-9}$	$1.75 \cdot 10^{-9}$	$2 \cdot 10^{-9}$

Table 3. Temperature dependences of the parameters characterizing dielectric properties of transformer oil with magnetic liquid impurity.

Temperature, K	308	313	318	323
σ_{AC} , $\text{Ohm}^{-1}\text{m}^{-1}$	$3.09 \cdot 10^{-7}$	$3.53 \cdot 10^{-7}$	$4.06 \cdot 10^{-7}$	$4.51 \cdot 10^{-7}$
τ , ms	61	47	44	39
W , nm	34	36	39	48
α	0.06	0.06	0.06	0.06

The values of the electrical conductivity σ_{AC} of the transformer oil with the magnetic liquid impurity at different temperatures as well as the temperature dependences of other parameters of the transformer oil with the magnetic liquid impurity are given in Table 3.

To get a clearer view of the temperature dependences of the above mentioned parameters, some of them are analyzed through a graphical representation.

Fig. 3 shows the values of ε' for the transformer oil (curve 1) and transformer oil with the magnetic liquid impurity (curve 2) in the coordinates $\lg \varepsilon' (10^3/T)$. Based on the analysis of the obtained data, the following conclusions can be drawn. First, the value of ε' for the transformer oil with the magnetic liquid impurity (curve 2) varies much more with temperature than in the case of the transformer oil without impurities. Second, the temperature dependence of ε' in the coordinates $\lg \varepsilon' (10^3/T)$ is almost linear (curve 1 in Fig. 3), while for the transformer oil with the magnetic liquid impurity it is nonlinear. We believe that this is caused by the fact that the magnetic impurity is polar. This point will be considered in more detail in our subsequent studies.

Fig. 4 shows temperature dependence of ε' in the coordinates $\lg \sigma_{AC} (10^3/T)$ for the transformer oil (curve 1) and transformer oil with the magnetic liquid impurity (curve 2).

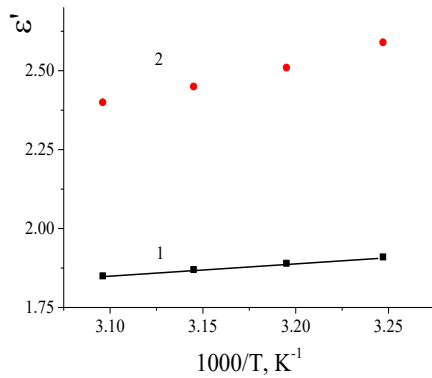


Fig. 3. Temperature dependences of the real part of the complex dielectric permittivity ε' for transformer oil without impurities (curve 1) and with magnetic liquid impurity (curve 2).

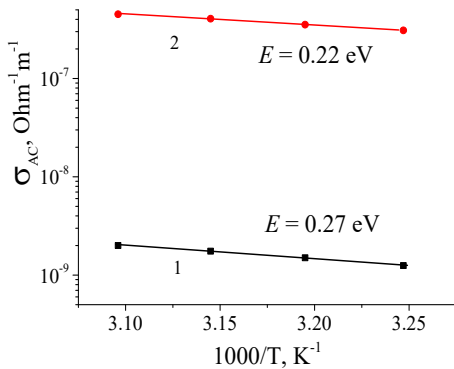


Fig. 4. Temperature dependences of electrical conductivity at alternating current σ_{AC} for transformer oil without impurities (curve 1) and with magnetic liquid impurity (curve 2).

It follows from analysis of Fig. 4 that the dependence $\lg \sigma_{AC} (10^3/T)$ is linear both in the case of the transformer oil without impurities (curve 1) and with the magnetic liquid impurity (curve 2). Mathematically, this dependence corresponds to the relation

$$\sigma_{AC} = \sigma_{\infty} \exp \frac{E}{kT}, \quad (2)$$

where σ_{∞} is the electrical conductivity at $T = \infty$, k is the Boltzmann constant, and E is the activation energy of the temperature dependence of electrical conductivity, respectively.

The values of the activation energy for the transformer oil (0.27 eV) and transformer oil with the magnetic liquid impurity (0.22 eV) are shown in Fig. 4. It can be concluded from the analysis of these values that introduction of magnetic impurities into the transformer oil leads not only to an increase in the electrical conductivity (by more than two orders of magnitude), but also to a decrease in the trap energy for the main charge carriers.

Finally, it is important to analyze the low-frequency relaxation process, which is observed in the transformer oil with the magnetic liquid impurity at the frequencies $f < 3 \cdot 10^3$ Hz. Analysis of such a process by using dielectric spectroscopy methods [7] showed that it can be described by the Cole–Cole relation (dispersion with a Gaussian distribution of relaxation times in magnitude). For this case, the relation between the components of the complex dielectric permittivity has the following form [9]:

$$\varepsilon^* = \varepsilon_{\infty} + \frac{\varepsilon_s - \varepsilon_{\infty}}{1 + (i2\pi f\tau)^{1-\alpha}}, \quad (3)$$

where ε^* is the complex permittivity, ε_{∞} is the value of permittivity at $f = \infty$, ε_s is the value of permittivity at $f = 0$, and α is the Cole–Cole parameter, respectively. The value of the parameter α can vary from 0 to 1.

The most of the values of the parameters in the relation (3) are given in Table 3. It follows from this table that the time of the relaxation process described by the relation (3) is tens of microseconds and decreases with temperature. This value of the relaxation time is close to the value of relaxation time τ at room temperature obtained in [7]. The value of the Cole–Cole parameter α coincides with the data for one of these two samples given in [7].

Separately, we analyze the temperature dependence of the thickness of the near-electrode region, where changes causing manifestation of the relaxation process described by the relationship (3) occur. To estimate this thickness W , we used the following relationship [7]:

$$W = \frac{d\varepsilon_{\infty}}{2\varepsilon_s}, \quad (4)$$

where d is the cell thickness. The values of W as well as other parameters of the samples are listed in Table 3. Analysis of the obtained data allows one to conclude that the value of W increases with temperature, similar to the value of σ_{AC} .

4. Conclusions

The dielectric properties of transformer oil without impurities and with magnetic liquid impurity were investigated in the frequency range of 6 to $7 \cdot 10^5$ Hz and at the temperatures of 308 to 423 K. The concentration of the impurity was 4.2% by volume. It is shown that:

1. The dielectric constant of the transformer oil ϵ' increases almost linearly with temperature T in the range of 308 to 423 K. Such behavior of ϵ' and its value of 1.9 makes it possible to assert that the electronic component is the main component of polarization of the transformer oil without impurities.

2. The dielectric constant of the transformer oil with the magnetic liquid impurity with a concentration of 4.2 vol.% at a temperature of 308 K is 1.36 times higher than the dielectric constant of the transformer oil without impurities. This difference increases with temperature. It leads to a conclusion that the magnetic impurity is polar.

3. The electrical conductivity of the transformer oil without impurities in the entire frequency range and the electrical conductivity of the transformer oil with the magnetic liquid impurity with a concentration of 4.2 vol.% at the frequencies $f > 3 \cdot 10^3$ Hz at a specific temperature do not depend on the frequency. These values reflect the bulk properties of the liquid. It is demonstrated that the temperature dependence of the electrical conductivity values corresponds to the Arrhenius law. It is found out that the activation energy of the temperature dependence of the electrical conductivity in the case of pure transformer oil is equal to 0.27 and 0.22 eV in the case of the presence of the magnetic impurity with the concentration of 4.2 vol.%. Such a difference in the activation energy values can most likely be caused by the fact that the magnetic impurity creates traps for the majority charge carriers with smaller energy depth than those existing in pure transformer oil.

4. The dispersion of the values of the real ϵ' and imaginary ϵ'' components of the complex dielectric permittivity for the transformer oil with the magnetic liquid impurity corresponds to the Cole–Cole dispersion, as was shown earlier in [7]. Based on the analysis of the Cole–Cole diagrams (dependences of ϵ'' on ϵ'), the values of the relaxation time τ , the thickness of the near-electrode region W , and the parameter characterizing the dispersion of relaxation times α are determined for each temperature. It is shown that the value of τ decreases with temperature, the value of W increases with temperature, and the value of α is equal to 0.06 and has no temperature dependence.

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Kopčanský P.: project administration, investigation, resources, software, writing – review & editing.

Температурні залежності параметрів, які характеризують діелектричні властивості трансформаторної олії з домішками магнітної рідини

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Анотація. У діапазоні частот від 6 до $7 \cdot 10^5$ Гц і температур 308...423 К досліджено діелектричні властивості чистої трансформаторної олії, а також трансформаторної олії з домішкою магнітної рідини з концентрацією 4,2% за об'ємом. Показано, що діелектрична проникність трансформаторної олії майже лінійно збільшувалась від температури. Діелектрична проникність трансформаторної олії з домішкою магнітної рідини за величиною перевищувала діелектричну проникність трансформаторної олії без домішок і нелінійно залежала від температури. Показано, що електропровідність трансформаторної олії без домішок у всьому діапазоні частот і електропровідність трансформаторної олії з домішкою магнітної рідини при частотах $f > 3 \cdot 10^3$ Гц за конкретної температури не залежали від частоти. Ці величини електропровідності відображають об'ємні властивості рідини. Показано, що температурна залежність цих величин електропровідності відповідає арреніусівському закону. Знайдено, що енергія активації для температурної залежності електропровідності у випадку чистої трансформаторної олії дорівнює 0,27 еВ, а у випадку наявності магнітної домішки – 0,22 еВ. Показано, що дисперсія величин дійсної та уявної компонент комплексної діелектричних проникностей для трансформаторної олії з домішкою магнітної рідини відповідає дисперсії Коул–Коула. На основі аналізу діаграм Коул–Коула для кожної із температур зразків знайдено величини часу релаксації τ , товщини приелектродної ділянки W та параметра, який характеризує дисперсію часів релаксації α . Показано, що величина τ зменшується з ростом температури, величина W збільшується з ростом температури, а величина α становила 0,06 і від температури не залежала.

Ключові слова: діелектрична проникність, електропровідність, час релаксації, трансформаторна олія, домішка магнітної рідини.