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Hetero- and low-dimensional structures

Dielectric properties of Shell transformer oil with impurities of carbon nanotubes and fullerene C_{60}

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Abstract. At the temperature 293 K, the influence of two types of nanoimpurities (carbon multiwall nanotubes and C_{60} fullerene) both separately and together on the dielectric properties of Shell transformer oil has been studied. It has been shown that these impurities do not significantly effect on the value of the dielectric permittivity of Shell oil, but more significantly increase its conductivity. It has been found that in the presence of nanotubes inside Shell oil, the dependence of its electrical conductivity on the fullerene concentration is nonmonotonic. The samples with the fullerene concentration 100 ppm have the highest conductivity. At the fullerene concentration 300 ppm, the conductivity of Shell oil with the impurities of carbon nanotube and C_{60} fullerene becomes almost equal to the electrical conductivity of Shell oil only with the impurities of carbon nanotubes. It has been suggested that C_{60} fullerene can be used to reduce the electrical conductivity of Shell oil with magnetic nanoparticles required to increase the cooling efficiency of transformers under the action of their own magnetic field.

Keywords: carbon multiwall nanotubes, fullerene C_{60}, transformer oil, dielectric permittivity, electrical conductivity.

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1. Introduction

It is known that when transmitting electrical energy over long distances, transformers are used. When an electric current passes through the windings of transformers, Joule heat is released, which must be transferred to the environment with a maximum efficiency to avoid the effect of overheating the transformers. Overheating of transformer windings can significantly reduce their service life.

Physical properties of the magnetic fluids (ferrofluids) have been studied by numerous research groups as many technical applications were uncovered. Especially, transformer oil-based ferrofluids in consequence of their enhanced thermal properties and higher breakdown voltage, as compared to pure oil, induced very intensive study of these materials. The magneto-dielectric effect what means the dependence dielectric permittivity on the applied magnetic field was studied by various authors [1]. The dispersion of magneto-dielectric anisotropy as a function of the electric field frequency and its dependence on the value of the magnetic field strength were presented in [2, 3]. To explain the low frequency relaxation process, the Schwarz model was applied [4], where the fact that colloidal nanoparticles are electrically charged by fixed or adsorbed ions and surrounded by counterions. So, they form a double electric layer on the surface of each particle, and it is under consideration. On the other hand, the position of frequency relaxation maximum shifts towards higher frequencies with increasing the magnetic volume of nanoparticles and the corresponding number of double layers. It was also proved [5, 6] that the low frequency relaxation process is slowed down, when direct current bias field is applied to the sample, which was related to formation of aggregates. In earlier studies [3, 7] on transformer oil based ferrofluids, the low-frequency relaxation process was assigned to double electric layer polarization on the particle surfaces.

To increase the cooling efficiency of transformer windings produced by industry, they are placed inside transformer oil [8]. In this case, cooling the windings occurs due to the presence of convection flows in this liquid.

To accelerate the flow rate of the liquid and thus increase the cooling efficiency of the transformer windings, in [9] it was proposed to add magnetic nanoparticles to the transformer oil. As we showed in [10], magnetic impurities significantly increase the electrical conductivity of transformer oil. This effect largely depends on the presence of various types of impurities in the transformer oil. Increasing the electrical conductivity of transformer oil due to introduction of conductive impurities together with the magnetic nanoparticles can lead to a decrease in the efficiency of the transformer due to the transfer of currents through the transformer oil. In addition, the presence of these currents can lead to additional (as compared to the existing one) heating the transformer oil and reduce the efficiency of its cooling caused by more intense fluid flows stimulated by the action of a magnetic field.

Therefore, the urgent problem is to find such impurities (of a separate type or several types together) that would be introduced into the transformer oil to reduce its electrical conductivity. As we have shown in [11, 12], a particular type of nanoparticles can reduce the electrical conductivity of liquid crystal due to adsorption properties. The task, from the practical and scientific viewpoint, is to investigate whether it is possible to reduce the electrical conductivity of transformer oil by adding nanoparticles of not one type, but at least two types. At the initial stage of these studies, it was decided to use multilayer carbon nanotubes as an active impurity instead of magnetic nanoparticles.

The aim of this work was to investigate the effect of C_{60} fullerene on the dielectric properties of transformer oil with impurities of carbon nanotubes.

2. Materials and methods of research

As in [10], we used transformer oil by the firm Shell for research. In addition to the Shell oil without impurities, Shell oil with impurity of fullerene C_{60} (the impurity concentration was 100 ppm), Shell oil with impurity of multilayer carbon nanotubes (MNTs) with the concentration 0.01 wt.%. Shell oil with the impurity of MNTs with the concentration 0.01 wt.% and impurity of fullerene C_{60} was investigated, too. In these studies, the concentration of C_{60} fullerene varied within the range from 0 up to 300 ppm.

To research dielectric properties, we used the same type of measuring cells as in [10]. The thickness of the cells was 20 μm. The cells were filled using capillary forces, because the transformer oil without/with the investigated nanoparticles well enough moistened the glass plates, from which the measuring cells were made. Measurements of the dielectric properties were performed using the oscilloscopic method at the temperature 293 K [13]. The amplitude of the measuring signal with sinusoidal shape was 5 V. The frequency range, at which the measurements were carried out, was from 6 to $5 \cdot 10^5$ Hz.

In the analysis of the obtained oscillograms, taking the resistance *R* and capacitance *C* connected in parallel as the sample scheme, we determined the values of resistance and capacitance of the tested samples, respectively. The value of conductivity for the alternating current σ_{AC} was determined using the *R*-value and known cell sizes, while the dielectric permittivities ε of Shell oil without/with impurities of fullerene as well as multiwall nanotubes were determined by the value of capacitance and known values of geometric dimensions of the measuring cell.

3. Results and discussion

Analysis of the obtained data showed that within the frequency range from 6 to $5 \cdot 10^5$ Hz, the ε-value did not depend on frequency. The values of ε for both pure Shell oil and Shell oil with impurities of MNTs and fullerene C_{60} both separately and together are listed in the table.

As can be seen from the table, C_{60} nanoparticles have virtually no effect on the ε-value of Shell oil. A very small increase in the ε-value for the C_{60} concentrations 200 and 300 ppm practically does not exceed the measurement error of this value (0.01). More significantly (as follows from the table) the nanotubes influence on the ε-value. From geometric considerations, this is understandable, because fullerenes are spherically symmetrical, and nanotubes, on the contrary, have no spherical symmetry.

Table. Influence of multiwall carbon nanotubes and fullerene C_{60} on the electrical properties of Shell transformer oil.

Sample	ε	σ_{AC}, $\text{Ohm}^{-1}\text{m}^{-1}$ (*f* = 1500 Hz)
Shell oil	1.91	$2.6 \cdot 10^{-8}$
Shell oil+ 0.01 wt.% MNTs	2.20	$4.6 \cdot 10^{-8}$
Shell oil + 100 ppm C_{60}	1.91	$2.7 \cdot 10^{-8}$
Shell oil + 0.01 wt.% MNTs + 100 ppm C_{60}	2.20	$4.4 \cdot 10^{-8}$
Shell oil + 0.01 wt.% MNTs + 200 ppm C_{60}	2.21	$4.0 \cdot 10^{-8}$
Shell oil + 0.01 wt.% MNTs + 300 ppm C_{60}	2.22	$2.8 \cdot 10^{-8}$

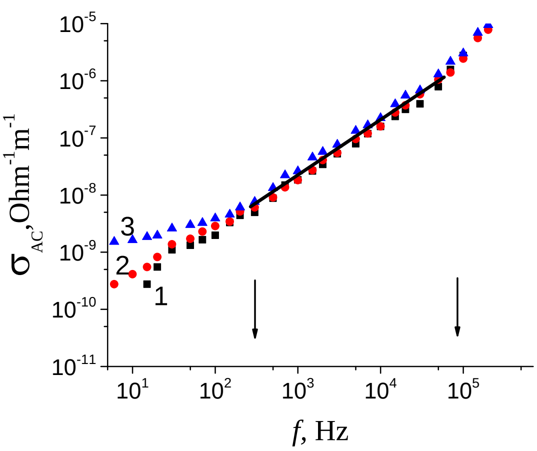

Fig. 1. Frequency dependences of conductivity for the alternating current σ_{AC} describing Shell oil (*1*), Shell oil + 100 ppm C_{60} (*2*) and Shell oil + 0.01 wt.% MNTs (*3*) at the temperature 293 K. The sample thickness is 20 µm.

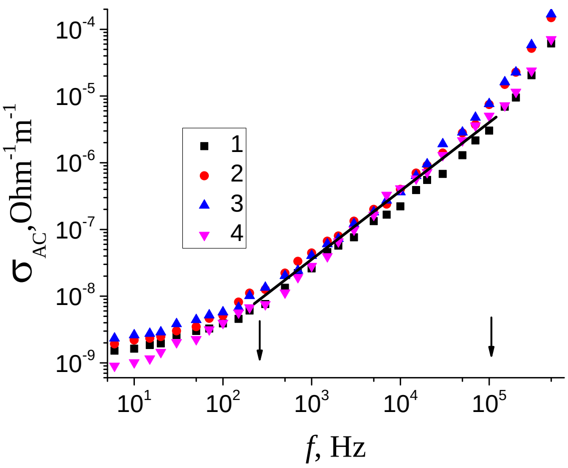

Fig. 2. Frequency dependences of conductivity for the alternating current σ_{AC} related to Shell oil + 0.01 wt.% MNTs at the various concentrations of C_{60} nanoparticles: 0 (*1*), 100 (*2*), 200 (*3*) and 300 ppm (*4*). Temperature 293 K. The thickness of the samples is 20 µm.

As our studies have shown, in contrast to the dielectric permittivity, the electrical conductivity of Shell oil depends much more on the presence of the nanoparticles of fullerene and multiwall carbon nanotubes than the dielectric permittivity ε. Another important difference was that, in contrast to the ε-value, the conductivity of both pure Shell oil and that with impurities of carbon nanotubes and fullerene C_{60} depends on frequency of the measuring signal.

Fig. 1 shows the frequency dependences of the conductivity for the alternating current σ_{AC} for Shell oil (curve *1*), Shell oil + 100 ppm C_{60} (curve *2*) and Shell oil + 0.01 wt.% MNTs (curve *3*). For a more unambiguous analysis, the obtained frequency dependences are separated into 3 sections by vertical arrows. The most significant difference for the conductivity of these samples is observed (as shown in Fig. 1) for the frequencies $f \leq 100$ Hz. As we showed in [14], at these frequencies the influence of near-electrode processes is significant. To analyze these processes in detail, it is necessary to perform measurements at lower frequencies. This may be the purpose of our further research.

From Fig. 1, we can also draw an important conclusion that the conductivity σ_{AC} on the second (by the *f*-value) section of the frequency range $10^2 \leq f \leq 10^5$ Hz in the double-logarithmic scale increases linearly with increasing the frequency. This means that the value of conductivity is a power function of frequency, *i.e.*, is described by the relationship:

$$\sigma_{AC} = k f^n,$$

where n is the exponent and k – parameter.

From the performed analysis of the frequency dependences on σ_{AC}, it follows that for the frequency dependences related to Shell oil, Shell oil + 100 ppm C_{60} and Shell oil + 0.01 wt.% MNTs the value $n = 1$. It means that the conductivity increases linearly with increasing the frequency of the measuring signal.

As it follows from Fig. 1, for $f \geq 10^5$ Hz the frequency dependence of conductivity differs from that observed for the section $10^2 \leq f \leq 10^5$ Hz. It may be caused by the fact that this is the area of frequencies where there is a transition to a purely electronic component of electrical conductivity (dipoles of molecules have no time to return during a change in the electric field strength that is equal to ¼ period of alternating electric field). With regard to the research performed in this paper, it is important to ascertain how the frequency dependences of the conductivity inherent to Shell oil will change in the presence of two types of impurities (fullerene and nanotubes). The frequency dependences of the conductivity for the alternating current σ_{AC} for Shell oil + 0.01 wt.% MNTs at different concentrations of nanoparticles C_{60} are shown in Fig. 2. Like to the case of the data shown in Fig. 1, there are 3 sections (in Fig. 2 they are marked with vertical arrows).

Since above we did not analyze in detail the frequency sections for $f \leq 100$ Hz and $f \geq 10^5$ Hz, indicating only the mechanisms by which they are caused, it makes no sense to do so for the data shown in Fig. 2. Therefore, we will analyze in more detail the results for the frequency range $10^2 \leq f \leq 10^5$ Hz. As follows from our analysis, the frequency dependence of the conductivity in this section is described by the same relation ($\sigma_{AC} = k f^n$) like to the data shown in Fig. 1.

Whereas this frequency dependence of conductivity is also characteristic for Shell oil without impurities, it can be concluded that the conductivity mechanism is set by Shell oil, and the introduced impurities only slightly affect its parameters. It is known from [15] that in single-component liquids the electrical conductivity does not depend on frequency. Even very well-purified Shell oil is not a single-component liquid [16]. This is the main reason that in this liquid the conductivity depends on frequency, which is characteristic for hop-like charge transfer in chemically heterogeneous media.

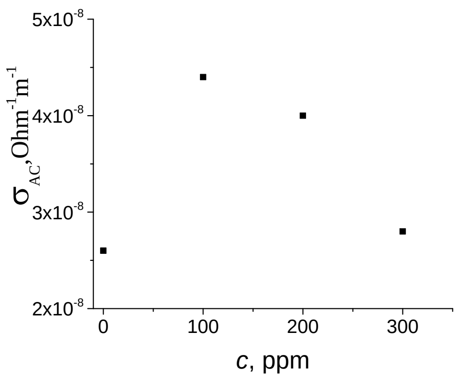

Fig. 3. Dependence of the electrical conductivity of Shell oil + 0.01 wt.% MNTs on the concentration of fullerene C_{60} for the frequency of the measuring signal 1500 Hz.

A detailed analysis of the data in Fig. 2 shows that the electrical conductivity of Shell oil + 0.01 wt.% MNTs is not a linear function of the fullerene C_{60} concentration. The dependence of the electrical conductivity of Shell oil + 0.01 wt.% MNTs on the concentration of fullerene C_{60} is shown in Fig. 3. From the obtained data, it is possible to draw a quite important conclusion that at the lowest of the selected fullerene concentrations (100 ppm), the electrical conductivity of the mixture Shell oil + 0.01 wt.% MNTs + 100 ppm C_{60} becomes higher than that of Shell oil + 0.01 wt.% MNTs, and then with increasing the concentration of C_{60} it begins to decrease, so that at the maximum (for research data) concentration of C_{60} it becomes almost equal to the electrical conductivity of Shell oil + 0.01 wt.% MNTs.

This can be explained by the presence of two mechanisms of the fullerene C_{60} influence on the electrical conductivity of Shell oil + 0.01 wt.% MNTs, namely: an increase in electrical conductivity due to introduction of new charge carriers (additionally to the existing ones in Shell oil + 0.01 wt.% MNTs) and a decrease in electrical conductivity due to their adsorption by fullerene C_{60}. At the concentrations of fullerene C_{60} less than 100 ppm, the first process prevails, and at the concentrations higher than 100 ppm – the second one does.

Being based on the obtained data, we can conclude that by varying the type of nanoparticles and their concentration, for Shell oil one can select such impurities that it would be possible to reduce the electrical conductivity of this liquid with magnetic nanoparticles and thus enhance the cooling effect of transformers due to additional flows to the convective ones in Shell oil owing to action of a magnetic field.

4. Conclusions

At the temperature 293 K, the effect of two different types of impurities (multiwall carbon nanotubes (MNTs) with the concentration 0.01 wt.% and fullerene C_{60}, the concentration of which varied within the range of 100 to 300 ppm) on the dielectric properties of Shell transformer oil has been studied.

1. It has been shown that only fullerene C_{60} with the concentration 100 ppm does not effect on the dielectric permittivity of Shell oil and increases its conductivity by only 4%.
2. Only MNTs with the concentration 0.01 wt.% increase the dielectric permittivity of Shell oil by 15%, and the electrical conductivity by 1.8 times.
3. It has been found that, within the frequency range 10^2 to 10^5 Hz, the electrical conductivity of Shell oil without/with impurities of fullerene and nanotubes varies according to the linear law of frequency. It has been assumed that such a conduction mechanism is caused by the properties of Shell oil and can be explained by the complex chemical composition of this liquid, since the most single-component liquids are characterized by the independence of electrical conductivity on frequency.
4. It has been shown that the electrical conductivity of Shell oil + 0.01 wt.% MNTs with fullerene nonmonotonically depends on the concentration of fullerene C_{60}. The maximum increase in conductivity by 1.7 times is observed at the concentration of fullerene C_{60} 100 ppm and further decreases with increasing the fullerene concentration, so that at its concentration 300 ppm the conductivity of Shell oil + 0.01 wt.% MNPs + 300 ppm fullerene C_{60} becomes almost equal to the electrical conductivity of Shell oil + 0.01 wt.% MNTs.

   It has been assumed that this dependence of conductivity is caused by two types of competing processes concerning the effect of fullerene C_{60} on the electrical conductivity of Shell oil + 0.01 wt.% MNTs. At the fullerene concentrations less than 100 ppm, the process of increasing the electrical conductivity due to introduction into Shell oil + 0.01 wt.% MNTs of additional charge carriers to the existing ones prevails in this liquid. At the fullerene concentrations higher than 100 ppm, the process of reducing the electrical conductivity due to adsorption of charge carriers by fullerene C_{60}, which are contained in Shell oil, prevails.
5. It has been proposed to use the mechanism of reducing the conductivity due to adsorption of charge carriers on the introduced impurities to reduce the conductivity of Shell oil with magnetic nanoparticles.

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Діелектричні властивості трансформаторної олії Shell з домішками вуглецевих нанотрубок та фулерену C_{60}

О.В. Ковальчук, І.П. Студеняк, Т.М. Ковальчук, E.A. Ayryan, K. Paulovičová, M. Timko, P. Kopčanský

Анотація. При температурі 293 K досліджено вплив двох типів нанодомішок (вуглецевих багатостінних нанотрубок та фулерену C_{60}) як окремо, так і разом на діелектричні властивості трансформаторної олії Shell. Показано, що такі домішки несуттєво впливають на величину діелектричної проникності трансформаторної олії, а більш суттєво збільшують її провідність. Знайдено, що при наявності у трансформаторній олії нанотрубок залежність її електричної провідності від концентрації фулерену є немонотонною. Найбільшу величину провідності мають зразки з концентрацією фулерену 100 ppm. При концентрації фулерену 300 ppm провідність трансформаторної олії з домішками вуглецевих нанотрубок та фулерену C_{60} стає майже рівною електропровідності трансформаторної олії з домішкою тільки вуглецевих нанотрубок. Зроблено припущення, що фулерен C_{60} можна використати для зменшення електропровідності трансформаторної олії з магнітними наночастинками, необхідними для збільшення ефективності охолодження трансформаторів під дією магнітного поля трансформатора.

Ключові слова: вуглецеві багатостінні нанотрубки, фулерен C_{60}, трансформаторна олія, діелектрична проникність, електропровідність.