Semiconductor Physics, Quantum Electronics & Optoelectronics, 2021. V. 24, No 2. P. 154-159.

Hetero- and low-dimensional structures

Influence of magnetic nanoparticles on dielectric properties of Shell oil transformer oil

O.V. Kovalchuk[1,2,3*], O.B. Nesterenko[1], V.Yo. Kotovskyi[2], I.P. Studenyak[4], T.M. Kovalchuk[5], K. Paulovičová[6], M. Timko[6], P. Kopčanský[6], K. Parekh[7], R.V. Upadhyay[7]

[1]*Kyiv National University of Technologies and Design,*
2, Nemirovich-Danchenko str., 01011 Kyiv, Ukraine
[2]*National Technical University of Ukraine "Igor Sikorsky Kyiv Polytechnic Institute"*
37, prospect Peremohy, 03056 Kyiv, Ukraine
[3]*Institute of Physics, NAS of Ukraine*
46, prospect Nauky, 03680 Kyiv, Ukraine
[4]*Uzhhorod National University, 46, Pidgirna str., 88000 Uzhhorod, Ukraine*
[5]*V. Lashkaryov Institute of Semiconductor Physics, NAS of Ukraine*
41, prospect Nauky, 03680 Kyiv, Ukraine
[6]*Institute of Experimental Physics, Slovak Academy of Sciences*
47, Watsonova str., 04001 Košice, Slovakia
[7]*Dr. K C Patel Research & Development Centre, Charotar University of Science & Technology,*
CHARSUAT campus, Changa-388421, Gujarat, India
[*]*Corresponding author e-mail: akoval@knutd.com.ua*

Abstract. The influence of two types of nano-impurities MF1 and MF2 on the dielectric properties of Shell oil transformer oil at the temperature 293 K has been studied. It has been shown that these magnetic impurities have no significant effect on the dielectric permittivity value of Shell oil, but more significantly increase its conductivity, in so doing, the impurity MF1 increases the conductivity of transformer oil 4 times larger than the impurity MF2. It has been ascertained that the low-frequency dielectric relaxation appearing in the studied samples can be described by the Cole–Cole equation. The parameters of this relaxation process and the influence of different types of magnetic impurities on them have been estimated.

Keywords: dielectric spectroscopy, dielectric relaxation, the Cole–Cole equation, magnetic nanoimpurities, dielectric properties.

https://doi.org/10.15407/spqeo24.02.154
PACS 75.30.Hx, 77.22.-d, 77.22.Gm

Manuscript received 20.04.21; revised version received 12.05.21; accepted for publication 02.06.21; published online 16.06.21.

1. Introduction

As widely known, the main source of energy on our planet is electricity. Currently, most of the world's electricity is generated by nuclear power plants (NPPs) and thermal power plants. Accidents at the Chernobyl nuclear power plant and, on a smaller scale, at the Japanese nuclear power plant in Fukushima have confirmed the need to build nuclear power plants in sparsely populated areas. This raises the problem of how to transmit electricity over long distances with minimal losses. In addition, for the stable operation of power engineering in a particular country or region, power plants are combined into a single energy system. This approach ensures stable operation even in the case when a particular power plant is in an emergency or is repaired, and it is disconnected from the power system. Of course, the transition to solar panels will create autonomous sources of electricity to some extent and thus reduces the amount of electricity needed for transmission. However, the cost of electricity generated by solar panels is still relatively high, which suggests that nuclear power plants and thermal power plants will be the main source of world electricity production for at least the next decades.

The main way to reduce losses in the transmission of electricity is to use the high voltage lines. Certainly, when the room temperature superconducting materials will be created, it will be significantly reduced the use of high-voltage power lines.

From the published data, it follows that the transition temperature to the superconducting state for recently synthesized materials is already approaching to room temperatures [1]. However, the cost of such materials is too high for their wide use for electricity transmission. Moreover, a record value for the transition temperature to the superconducting state was obtained with high compression of the material.

From the mentioned above, it can be concluded that at least in the nearest decades the important problem will be to reduce energy losses during the transmission.

In the existing power systems, the increase or decrease of voltage level is carried out using the transformer. Transformer oil is used to cool transformer windings, especially in the distribution line transformer and power transformer [2–9]. Heat transfer from the transformer windings to the environment is carried out by convection flows of the liquid. Since the operation of transformer produces sufficiently high magnetic fields, addition of magnetic impurities into the transformer oil can increase the cooling efficiency of the transformer by additional magnetic buoyancy force. However, such impurities can significantly affect the conductivity of transformer oil, which may lead to additional heating of the transformer oil. Therefore, increasing the cooling efficiency by enhancing the convective flow rate may not always serve the purpose when adding magnetic nanoparticles to the transformer oil.

From the analysis of the published data [2–9], it can be concluded that the effect of magnetic nanoparticles on the dielectric properties of transformer oil was not studied in detail. Therefore, the aim of this work is to study the effect of magnetic nanoparticles on the dielectric properties of transformer oil.

2. Materials and methods of research

The impurities MF1 and MF2 in the form of magnetic nanoparticles were prepared using co-precipitation method and dispersed in the transformer oil. In this work, we have used the transformer oil produced by the Shell Company (Shell oil). Both pure oil and oil with two magnetic impurities (MF1 and MF2) were investigated.

Since the conductivity of Shell oil is low even in the presence of magnetic nanoparticles, we have used the same type of measuring cells and measurement method as it was used for the study of liquid crystals [10]. The thickness of the cells was 20 μm. The filling of the cells took place in the same way as the filling of the liquid crystals by means of capillary forces, since the transformer oil without/with magnetic impurities moistens the glass surface quite well.

Measurement of the dielectric properties of the samples was performed using the oscilloscopic method [11] at the temperature 293 K. The amplitude of the measuring signal with sinusoidal shape was 5 V. The frequency range within which the measurements were made was 6 to $3\cdot10^5$ Hz.

In the analysis of the obtained oscillograms, taking as an equivalent sample scheme the resistance *R* and capacitance *C* connected in parallel, we determined, respectively, real ε' and imaginary ε'' components of complex dielectric permittivity for various frequencies of the measuring signal. Then the frequency dependences of ε' and ε'' were analyzed on the basis of existing theories of relaxation process as well as articles and monograph published by us [10, 12, 14, 15].

3. Results and discussion

Fig. 1 shows the frequency dependences of the real component of the complex dielectric permittivity ε' for the Shell oil without impurities (curve *1*), for Shell oil with the impurity MF2 (curve *2*) and Shell oil with the impurity MF1 (curve *3*).

From the analysis of Fig. 1 we can draw the following conclusions.

First, at the frequencies *f* higher than 200 Hz, the ε' value does not depend on frequency *f*. As we showed in [12], for this frequency range the influence of near-electrode processes is insignificant, and we can assume that the electric field is uniform along the thickness of the sample. The parameters of samples determined for these frequencies will correspond to the parameters that characterize their bulk properties.

One of these parameters is the relative dielectric permittivity ε_∞. The ε_∞ values for the studied samples are listed in Table.

From the analysis of the ε_∞ values given in the table, it can be concluded that when magnetic impurities are added to Shell oil, the value of the dielectric permittivity increases. Since the concentration of these impurities was low, in addition to the magnetic properties, the impurities are characterized by sufficiently high dielectric properties. Being based on the purpose of this work, the dielectric properties of magnetic impurities alone have not been studied.

Second, at the frequencies lower than 200 Hz, for Shell oil with magnetic impurities (curves *2* and *3* in Fig. 1), there is a dispersion of the ε' value depending on the frequency of the measuring signal. To determine the reason for this dispersion, the frequency dependences of the imaginary component of the complex dielectric permittivity ε'' were also investigated.

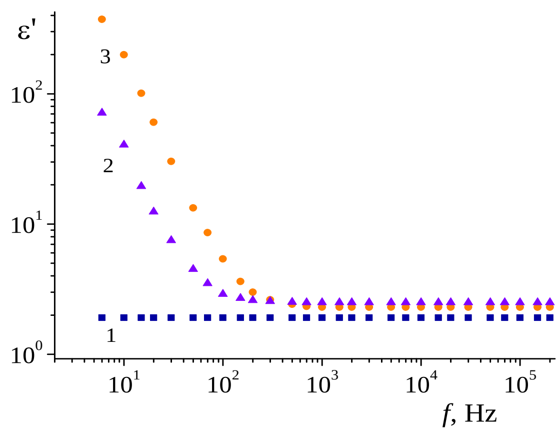

Fig. 1. Frequency dependences of the real component of the complex dielectric permittivity ε': Shell oil without impurities (*1*); Shell oil with the impurity MF2 (*2*) and Shell oil with the impurity MF1 (*3*). The thickness of the sample is 20 μm. The temperature for measuring the samples was 293 K.

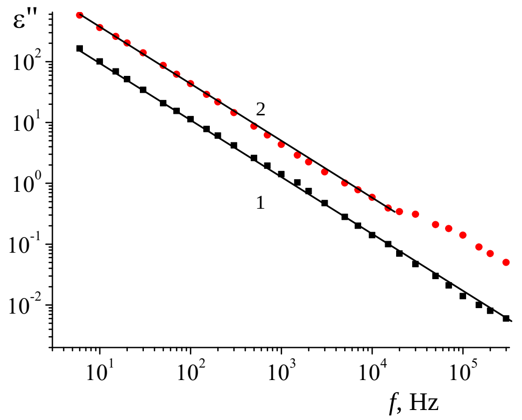

Fig. 2. Frequency dependences of the imaginary component of complex dielectric permittivity ε": Shell oil with the impurity MF2 (*1*) and Shell oil with the impurity MF1 (*2*). The sample thickness was 20 µm. The temperature of studied samples was 293 K.

The frequency dependences of the imaginary component of the complex dielectric permittivity ε" are shown in Fig. 2. As known, the imaginary component of the complex dielectric permittivity ε" is related with the conductivity of the sample by the relation:

$$\sigma = 2\pi f \varepsilon_\infty \varepsilon'' , \quad (1)$$

where σ is the specific electrical conductivity and ε_∞ is the dielectric permittivity for $f \to \infty$.

We have used just the relation (1) to determine the σ value for the Shell oil with magnetic nanoparticles. In the case of pure Shell oil, the conductivity was lower than the sensitivity of the device used for these measurements (10^{-10} $Ohm^{-1}\cdot m^{-1}$). Therefore, in Fig. 2 the data for the frequency dependences of ε" for Shell oil are not given and Table does not show the conductivity of pure transformer oil. As for the conductivity of Shell oil with magnetic impurities, it should be noted that the conductivity of transformer oil with the impurity MF1 is 4 times higher than that of oil with the impurity MF2.

Analyzing the frequency dependences of ε" shown in Fig. 2, it should be noted the deviation from the linear dependence on a two-logarithmic scale for Shell oil with the impurity MF1 for the frequencies lower than $2\cdot10^4$ Hz. The reason for this is obviously dipole polarization of impurities MF1. But these changes are much smaller than the dispersion of the components of the complex dielectric permittivity at the frequencies lower than 200 Hz. That is why just the reasons for the low-frequency dispersion in this paper were analyzed in more detail.

The dielectric properties of several types of transformer oil grades were studied in detail in the work [8]. It was shown that the frequency dependences of the dielectric permittivity components ε' and ε" can be described on the basis of the relation:

$$\varepsilon^* = \varepsilon_\infty + \frac{\varepsilon_{s1}}{1+(i2\pi f\tau_1)^{\alpha_1}} + \frac{\varepsilon_{s2}}{1+(i2\pi f\tau_2)^{\alpha_2}} + \frac{\sigma}{i\varepsilon_0 2\pi f} + \frac{b}{\varepsilon_0 (i2\pi f)^c} \quad (2)$$

where ε* is the complex dielectric permittivity; ε_s is the dielectric permittivity at $f = 0$; τ_1 and τ_2 are the dielectric relaxation times; α_1, α_2, *b* and *c* are constants.

Our analysis of the obtained experimental results showed that the relation (2) needs not to be used to describe the dielectric spectra. To ascertain the mechanism of low-frequency dispersion of the components inherent to the complex dielectric permittivity ε' and ε", the dependences ε"(ε') (Cole–Cole diagrams) were plotted and analyzed. The analysis of these diagrams showed that they can be described by an arc with a small error. As known [13], this relaxation process corresponds to the Cole–Cole dispersion and is described by the relation:

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

where α is the Cole–Cole parameter.

The α value can vary from 0 (Debye dispersion) to 1 (no dispersion) [13]. The values of α parameter for the studied samples with impurities are listed in Table. From the data obtained above, it follows that the α value is low (0.03 for the magnetic impurity MF2 Shell and 0.07 for the magnetic impurity MF1). It can be caused by the same composition of the impurity. Moreover, the impurity MF2 is more homogeneous in chemical composition than the impurity MF1. Obviously, it can explain the difference between the frequency dependences of ε" for Shell oil with impurities MF1 and those for Shell oil with impurities MF2 for the frequencies higher than $3\cdot10^4$ Hz (Fig. 2).

One of the main parameters characterizing the Cole–Cole dispersion is the dielectric relaxation time τ. According to the Table data, the τ value for Shell oil with the magnetic impurity MF2 is 1.2 s and 4 times higher than the corresponding value for Shell oil with the magnetic impurity MF1. That is, and in the case of transformer oil with these impurities, we observe the regularity that we found earlier, namely, the dielectric relaxation time that characterizes near-electrode processes is inversely proportional to the conductivity of the sample.

Another parameter of the relaxation process, which corresponds to the Cole–Cole dispersion and is described

Table. Parameters that characterize the bulk and near-electrode properties of Shell oil without/with the impurities MF1 and MF2.

Sample	ε_∞	ε_s	σ, $Ohm^{-1}\cdot m^{-1}$	τ, s	*W*, nm	α
Shell oil	1.9	–	–	–	–	–
MF1	2.5	1070	2.4 10^{-7}	0.39	23	0.07
MF2	2.3	430	7.8 10^{-8}	1.2	53	0.03

Notes. ε_∞ – dielectric permittivity for $f \to \infty$, ε_s is the dielectric permittivity for $f = 0$, σ is the specific electrical conductivity, τ is the dielectric relaxation time, *W* is the thickness of the near-electrode area, α is the Cole–Cole parameter.

by the relation (2), is the dielectric permittivity ε_s extrapolated to the frequency $f = 0$. If we assume that the parameters of the near-electrode areas close to each electrode are the same (which means the same capacitance of the near-electrode areas) and the dielectric permittivity of the near-electrode area is equal to ε_∞, then the thickness of the near-electrode area can be estimated using the maximum capacity of the sample for this relaxation process (obtained by analyzing the Cole–Cole diagram):

$$W = \frac{d}{2}\frac{\varepsilon_\infty}{\varepsilon_s}\,. \tag{4}$$

The thickness of the near-electrode areas W for Shell oil with various magnetic impurities estimated using the relation (3) is also given in Table. From these data, it can be concluded that, as in the case of relaxation time, the thickness of the near-electrode area is also inversely proportional to the conductivity. This conclusion confirms our previously obtained data for liquid crystals with impurities [14].

4. Conclusions

It has been studied the influence of two different types of magnetic nano-impurities MF1 and MF2, which are used to increase the cooling efficiency of transformers by increasing the rate of convection flows in the liquid under the magnetic field of these devices, on the dielectric properties of transformer oil Shell oil. The following inferences are drawn from the study.

1. It has been shown that magnetic nano-impurities do not significantly increase the dielectric permittivity of Shell oil (the impurity MF2 increases the dielectric permittivity by 20%, and the impurity MF1 – by 30%). However, the conductivity of Shell oil with introduction of the impurities increases much more. Moreover, in the case of introduction of impurities in Shell oil MF1, the conductivity of transformer oil increases to the value $2.4\cdot10^{-7}$ $\mathrm{Ohm^{-1}\cdot m^{-1}}$, which is 4 times higher than the conductivity of Shell oil with the same concentration of impurities MF2.

2. It has been determined that the low-frequency dispersion of the components of the complex dielectric permittivity (for frequencies lower than 200 Hz) of the studied samples can be described by the Cole–Cole equation. The dielectric relaxation time, the Cole–Cole parameter and the thickness of the near-electrode area, where the relaxation processes take place, have been estimated.

3. It has been determined that the dielectric relaxation time in the presence of the impurity MF2 in Shell oil is 1.2 s, which is more than 3 times higher than that in the presence of the impurity MF1 in transformer oil.

4. The thickness of the near-electrode area, where the relaxation process occurs, which is described by the Cole–Cole equation, has been estimated. In the case of the impurity MF2, this value equals 53 nm, and in the case of MF1 – 23 nm.

Acknowledgements

This work was supported by the Slovak Academy of Sciences and Ministry of Education in the framework of project VEGA 2/0011/20, Slovak Research and Development Agency under the Contract No. APVV-18-0160, project COST NANOConVEX CIG-15119 and project Structural Funds of EU MODEX "Modified (nano)textile materials for health technologies".

References

1. Snider E., Dasenbrock-Gammon N., McBride R. *et al*. Synthesis of yttrium superhydride superconductor with a transition temperature up to 262 K by catalytic hydrogenation at high pressures. *Phys. Rev. Lett.* 2021. **126**. P. 117003. https://doi.org/10.1103/PhysRevLett.126.117003.
2. Xiaobo Wang, Chao Tang, Bo Huang, Jian Hao. Review of research progress on the electrical properties and modification of mineral insulating oils used in power transformers. *Energies.* 2018, **11**. P. 487. https://doi.org/10.3390/en11030487.
3. Suhaimi N.S., Din M.F.Md, Rahman A.R.A. *et al*. Optimum electrical and dielectric performance of multi-walled carbon nanotubes doped disposed transformer oil. *Energies.* 2020. **13**. P. 3181. https://doi.org/10.3390/en13123181.
4. Dehkordi M.F. *Dielectric behavior of transformer oil when contaminated and/or fortified with nano-particles*. Master of Engineering dissertation. Université du Québec à Chicoutimi, Canada, 2014.
5. Hägerbrand J. *Measurements of resistivity in transformer insulation liquids*. Independent thesis Advanced level (professional degree), Master in Electrical Engineering. Uppsala Universitet, Department of Electrical Engineering. Sweden, 2020.
6. Hussain M.R., Refaat S.S. Dielectric behaviour of defects in power transformer insulation using finite element method. *2019 2nd Intern. Conf. on Smart Grid and Renewable Energy (SGRE)*, 2019. P. 1–6. https://doi.org/10.1109/SGRE46976.2019.9021069.
7. Hessien M.M., Sabiha N.A., Ghoneim S.M., and Alahmadi A.A. Enhancement of dielectric characteristics of transformer oils with nanoparticles. *Int. J. Appl. Eng. Res.* 2017. **12**, No 24. P. 15668–15673. https://doi.org/10.13140/RG.2.2.16376.34565.
8. Yang L., Gubanski S.M., Serdyuk Yu.V., and Schiessling J. Dielectric properties of transformer oils for HVDC applications. *IEEE Trans. on Dielectrics and Electr. Insulation*. 2012. **19**, No 6. P. 1926–1933. https://doi.org/10.1109/TDEI.2012.6396949.
9. Ten C.F., Fernando M.A.R.M. and Wang Z.D. Dielectric properties measurements of transformer oil, paper and pressboard with the effect of moisture and ageing. *2007 Annual Report – Conference on Electrical Insulation and Dielectric Phenomena*, 2007. P. 727–730. https://doi.org/10.1109/CEIDP.2007.4451521.
10. Poberezhets S.I., Kovalchuk O.V., Studenyak I.P. *et al.* Temperature dependence of dielectric properties

of the liquid crystal 6CB with the embedded Ag_7GeS_5I nanoparticles. *SPQEO*. 2020. **23**. P. 129–135. https://doi.org/10.15407/spqeo23.02.129.
11. Twarowski A.J., Albrecht A.C. Depletion layer in organic films: Low frequency measurements in polycrystalline tetracene. *J. Chem. Phys.* 1979. **70**. P. 2255–2261. https://doi.org/10.1063/1.437729.
12. Koval'chuk A.V. Generation of charge carriers and formation of antisymmetric double electric layers in glycerine. *J. Chem. Phys.* 1998. **108**. P. 8190–8194. https://doi.org/10.1063/1.476174.
13. Barsukov E., Macdonald J.R. *Impedance Spectroscopy. Theory, Experiment and Applications*. John Wiley & Sons, 2005.
14. Koval'chuk A., Dolgov L., Yaroshchuk O. Dielectric studies of dispersions of carbon nanotubes in liquid crystals 5CB. *SPQEO*. 2008. **11**, No 4. P. 337–341. https://doi.org/10.15407/spqeo11.04.337.
15. Kopčanský P., Timko M., Studenyak I.P., Kovalchuk O.V. *Nanoparticles in Homogeneous, Micro- and Nanometric Structures Based on Liquid Crystals: Morphology and Dielectric Properties*. Institute of Experimental Physics, Slovak Academy of Sciences, Košice, 2020.

Authors and CV

Oleksandr V. Kovalchuk, Doctor of Physical and Mathematical Sciences, senior scientific researcher at the Molecular Photoelectronics Department, Institute of Physics, NAS of Ukraine; professor of the Department of Applied Physics and High Mathematics at the Kyiv National University of Technologies and Design as well as professor at the National Technical University of Ukraine "Igor Sikorsky Kyiv Polytechnic Institute". Authored over 150 articles, 8 patents. The area of his scientific interests is dielectric spectroscopy of liquid crystals and composites.

Olga B. Nesterenko, PhD in Physics and Mathematics, Associate Professor, Head of the Department of Applied Physics and Higher Mathematics at the Kyiv National University of Technologies and Design. Author of more than 60 publications. The area of her scientific interests is the research of the problems of natural and applied sciences with mathematical methods.

Tetiana M. Kovalchuk, scientific researcher at the V. Lashkaryov Institute of Semiconductor Physics, NAS of Ukraine. Authored over 20 articles. The area of her scientific interests is dielectric spectroscopy.

Vitalii Yo. Kotovskyi, Doctor of Technical Sciences, Professor, Head of the Department of General Physics at the National Technical University of Ukraine "Igor Sikorsky Polytechnic Institute". He is the author of more than 193 scientific publications. His main research interests are solid state physics, modern information and energy saving technologies.

Ihor P. Studenyak, defended his Dr. Sc. degree in Physics and Mathematics in 2003 and became full professor in 2004. Vice-rector for research at the Uzhhorod National University, Ukraine. Authored over 200 publications, 120 patents, 15 textbooks. The area of his scientific interests includes physical properties of semiconductors, ferroics and superionic conductors.

Katarína Paulovičová, is working at the Department of Magnetism of Institute of Experimental Physics SAS, Košice since 2018 as a young scientific researcher. She received her PhD at the Technical University of Košice. Her interest is devoted to the synthesis and basic characterization of magnetic nanoparticles and magnetic nano-fluids based on the electrical insulating fluids for electrical engineering applications. In addition, her research activities are focused on the study of the rheological behavior of prepared nanofluids in the flow and experimental study of change rheological properties of this nanofluids affected by external applied magnetic or electric field.

Milan Timko, PhD in solid state physics. Senior researcher of Institute of Experimental Physics, Slovak Academy of Science. Authored over 220 articles, 4 patents, 1 monograph, and 3 text-books. The area of his scientific interests includes solid state physics, magnetic fluids and their magnetic, dielectric and hyperthermia properties.

Peter Kopčanský, Professor in solid state physics. Senior researcher of Institute of Experimental Physics, Slovak Academy of Sciences. Authored over 250 articles, 6 patents, 1 monograph, 5 textbooks. The area of his scientific interests includes solid state physics, especially magnetism, transport properties in disordered systems, magnetic fluids, their magnetic and dielectric properties and composite systems with liquid crystals.

Kinnari H. Parekh, Senior Research Scientist, has received her PhD degree in Physics in 1999 from Bhavnagar University. She was an assistant professor at M S University of Baroda (2003 to 2009) and then at Indian Institute of Technology Gandhinagar (2008 to 2011). From 2011 onwards she has joined Charotar University of Science & Technology, Changa. She has published more than 80 papers in international journals, 2 patents, 1 book chapter published by the CRC Press, more than 80 presentations at international conferences. About 1070 citations (WOS), h = 17. She works on the application oriented synthesis and characterization of magnetic fluids for engineering and biomedical fields. Specialist in large scale production of magnetic fluids. She is a recipient of BOYSCAST award and DST young scientist award in terms of FTPYS project.

Ramesh V Upadhyay, Professor, received his PhD from Saurashtra University, Rajkot, India in the year 1985. He has published more than 175 papers in International journals, 2 book chapters; more than 100 presentations at National and International conferences. He was a Commonwealth Academic Staff fellow at the University of North Wales, Bangor during 1991-1992. He was a STINT-Visiting Professor at KTH, Sweden. At present, major thrust area of research is focused on synthesis of MR and magnetic fluids for industrial applications like, dampers, gearbox coolant, *etc*. He has 4 patents to his credit.

Вплив магнітних наночастинок на діелектричні властивості трансформаторної олії Shell oil

О.В. Ковальчук, О.Б. Нестеренко, В.Й. Котовський, І.П. Студеняк, Т.М. Ковальчук, K. Paulovičová, M. Timko, P. Kopčanský, K. Parekh, R.V. Upadhyay

Анотація. Досліджено вплив двох типів нанодомішок MF1 та MF2 на діелектричні властивості трансформаторної олії Shell oil при температурі 293 K. Показано, що такі магнітні домішки несуттєво впливають на величину діелектричної проникності Shell oil, проте більш суттєво збільшують її провідність, причому домішка MF1 збільшує провідність трансформаторної олії у 4 рази більше, ніж домішка MF2. Встановлено, що низькочастотну діелектричну релаксацію, яка виникає у досліджуваних зразках, можна описати рівнянням Коул–Коула. Оцінено параметри такого релаксаційного процесу і вплив на них різного типу магнітних домішок.

Ключові слова: діелектрична спектроскопія, діелектрична релаксація, рівняння Коул–Коула, магнітні нанодомішки, діелектричні властивості.