

Peculiarities of the effect of different types of SOR nanoimpurities on the value of ionic component of the electrical conductivity of the homeotropically aligned nematic liquid crystal 6 CB

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Abstract. In this paper, the effects of SOR5, SOR10, and SOR15 nanoimpurities on the value of the ionic conductivity of the homeotropically aligned nematic liquid crystal 6CB are reported. Electrical measurements are carried out within a wide frequency range (from 6 Hz to 10⁶ Hz) at room temperature (293 K). The largest changes in the electrical conductivity occur in the low-frequency range (less than 10³ Hz) and depend on the type of nanoimpurity and on its concentration. Despite the similarity in the chemical composition of SOR5, SOR10, and SOR15, the measured dependences of the electrical conductivity of the studied samples on the concentration of nanoimpurities are substantially different. In the case of the SOR10 impurity, the ionic component of the electrical conductivity depends on the concentration of SOR10 according to a power law with an exponent approximately equal to 0.5, which is typical for the bimolecular recombination of charge carriers (*i.e.*, the behavior of a weak electrolyte). Liquid crystal samples containing SOR5 and SOR15 nanoimpurities behave in a similar way in a low concentration region (between 0.01 and 0.05 mass %). Interestingly, further increase in the concentration of nanodopants (>0.05 mass %) results in a dramatically different behavior. The electrical conductivity of liquid crystals doped with SOR5 undergoes a sharp increase whereas the electrical conductivity of samples containing SOR15 decreases.

Keywords: dielectric properties, nematic liquid crystal, homeotropic alignment of molecules, ionic conductivity, nanoimpurity concentration.

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1. Introduction

In our recent work [1], we investigated the effect of SOR5 nanoparticles on the dielectric properties of the homeotropically aligned nematic liquid crystal (LC) 6CB within the frequency range 6 to 10⁶ Hz. The measured frequency spectrum was separated into three regions. For each of these regions, conclusions were drawn regarding

the effects of nanoparticle concentration and cell thickness on the frequency dependence of the components of the complex dielectric permittivity on the frequency of the measuring signal. Because in addition to SOR5, the samples of liquid crystals doped with other nanoimpurities of similar chemical composition, in particular SOR10 and SOR15, were prepared at the Institute of Experimental Physics of the Slovak Academy of Sciences,

it was important to investigate how the type of impurity will affect the changes in the electrical conductivity of the same liquid crystal materials. One of the first questions that arise in such studies is the question of how much each of the impurities changes the electrical properties of the liquid crystal itself [2, 3]. Therefore, it was important to study exactly the region of the dielectric spectrum, where the presence of an impurity affects the LC conductivity.

It is known [2–5] that molecular liquid crystals have an ionic type of conductivity. In the works [2–5], it was shown in detail that purely ionic conductivity of liquids does not depend on the frequency. The same can be applied to molecular LC because they are anisotropic dielectric liquids. From a scientific viewpoint, it was important to uncover how this quantity could be estimated. In our previous works, it was shown that, under certain conditions, it is possible to define the regions of the dielectric spectrum where the LC conductivity even with various types of impurities does not depend on the frequency.

The simplest ways are to increase the thickness of the measuring cell and reduce the frequency of the measuring signal [1]. Even though the measuring device did not allow performing the research at very low frequencies, the search for regions of the dielectric spectrum, where the value of the dielectric permittivity did not depend on the frequency, was carried out on the basis of the analysis of the dielectric spectra obtained at various frequencies of the measuring signal.

Therefore, the purpose of this work was (i) to ascertain, through research at various thicknesses of the liquid crystal, under which conditions it is possible to define a region of the dielectric spectrum where the conductivity of the liquid crystal containing a specific impurity does not depend on the frequency (in the case of adding SOR10 and SOR15 impurities into 6CB), and (ii) to investigate the peculiarities of the influence of these impurities on the LC conductivity. Since this problem was not considered in the publication [1], it was addressed in this paper. That is, the main task of this work was to investigate how the SOR5, SOR10 and SOR15 impurities affect the ionic conductivity of 6CB. In the case of the SOR5 impurity, we took advantage of the results obtained in [1].

It was noted in [1] that the effect of the SOR5 impurity on the 6CB conductivity has not been studied before. The same is applied to the SOR10 and SOR15 impurities. That is, the study of the effects of the SOR5, SOR10, and SOR15 impurities on the ionic conductivity of nematic liquid crystal 6CB can be considered as the scientific novelty of the present paper. In previous works analyzed in detail in the reviews [3, 6], the possibility of different effects of nanoparticles on the ionic conductivity of molecular liquid crystals was noted. This work experimentally demonstrates three types of behavior inherent to the ionic conductivity of molecular liquid crystals doped with the SOR5, SOR10, and SOR15 nanoimpurities.

2. Research materials and methods

Like in the work [1], we used nematic liquid crystal 6CB. The measuring cells had a sandwich structure. For this type of cells, the conditions at the periphery of the sample can have a significant impact because of edge effects. Therefore, the measuring electrodes had two sections – central and peripheral. Just the central part of the sample was used as the measuring electrode. The peripheral part was used as a guard electrode (to eliminate edge effects). The electric current through this part of the sample did not affect the measurement process. As in [1], we used ITO electrodes (a mixture of tin and indium oxides) that are transparent in the visible range of the optical spectrum.

The homeotropic alignment of LC in the cell was achieved in the same way as described in [1]. Examinations of samples using a polarizing microscope showed that even at the maximum concentration of the impurity (0.1 mass %), the homeotropic orientation of LC molecules was not disturbed even in the absence of electric field. Since 6CB has a positive value of dielectric permittivity anisotropy, then external electric field was an additional stabilizing factor for the homeotropic alignment of LC molecules.

The research was performed using cells of three different thicknesses d (5, 20 and 50 μm). The thickness of the cell was controlled by polymer spacers that were applied to the guard electrodes. To reduce the influence of the environment, after filling the cell, it was sealed by applying epoxy glue around the perimeter of the sample.

In this work, we investigated LC 6CB doped with SOR5, SOR10, and SOR15 impurities by using three impurity concentrations: 0.01, 0.05 and 0.1 mass %. Even though some results describing the effect of SOR5 impurity on the ionic conductivity of 6CB were reported in our previous work [1], a comparative analysis of the effect of different types of SOR-based nanodopants on the ionic conductivity of liquid crystal 6CB is still missing.

The study of the dielectric properties of 6CB with SOR5 impurities was carried out using the oscilloscopic method [7, 8] and sinusoidal measuring signals. The amplitude of the measuring signal was 2.5 V. The dielectric properties of the samples were measured within the frequency range $f = 6 \dots 10^6$ Hz at the temperature 293 K. Using the oscilloscopic method, the values of electrical resistance R and electrical capacitance C were found for individual frequencies, assuming that the equivalent circuit of the sample is a resistor and a capacitor connected in parallel. To analyze frequency dependences, we used a logarithmic scale (since the frequency of the measurement signal varied within rather wide ranges). The frequencies of the generator were chosen so that the interval between them in the logarithmic scale was the same.

According to the known geometric dimensions of the studied samples, on the basis of the obtained values of resistance R and capacitance C of the samples,

the imaginary ε'' and real ε' components of the complex dielectric permittivity ε^* were obtained, respectively. As will be shown below, the main parameter for analysis in this work is the resistance of the sample, and the main meaning of this analysis is the conductivity value of the samples σ at various frequencies. The main conclusions of the present work were made by analyzing the frequency dependences of σ .

As it was already noted above, in this work it was necessary to investigate the effect of SOR5, SOR10 and SOR15 impurities on the ionic conductivity of LC 6CB. A characteristic feature of the manifestation of only ionic conductivity is the absence of dependence of conductivity on frequency [4, 5]. The performed studies using various thicknesses of LC as well as various concentrations of impurities showed that these dependences are observed at the lowest of the selected frequencies of the measuring signal and the largest of the selected thicknesses of the measuring cell (50 μm). Therefore, in the experimental part of this work, an analysis of the frequency dependences of the conductivity obtained using 50 μm thick cells is presented.

3. Experimental results and their analysis

Fig. 1 shows the frequency dependences of the real (ε') component of the complex dielectric permittivity of nematic liquid crystals 6CB containing the maximum concentration (0.1 mass %) of impurities SOR5 (curve 1), SOR10 (curve 2) and SOR15 (curve 3).

Similar to the analysis of dielectric spectrum reported in the work [1], three regions of the measured frequency dependence can be distinguished. There are two regions for $f < 10^2$ Hz and $f > 2 \cdot 10^5$ Hz, where changes in the value of ε' are observed with a change in the frequency of the measuring signal f , and the plateau region of the spectrum, where the value of ε' does not depend on the frequency. As it was noted in the works of other authors [9], as well as in our works [1, 8], changes in the value of ε' depending on the low frequencies (< 10 Hz) are caused by near-electrode processes. In this case, to ensure charge exchange between the electrode and LC, almost the entire voltage of the measuring signal will be applied across the near-electrode area. This leads to a significant increase in the effective value of ε' . Although, in fact, the value of ε' does not increase. This apparent increase is caused by the nonuniform distribution of the applied electric field across the sample thickness [8].

In the region of the dielectric spectrum for $f > 2 \cdot 10^5$ Hz, the decrease in the value of ε' with increasing the frequency is caused by dipole polarization. Dipoles of molecules have no time to return in a time interval close to the period of the measuring signal. This phenomenon is typical for the most liquids and will not be analyzed in detail.

In this work, the region of dielectric spectrum where the value of ε' does not depend on the frequency is

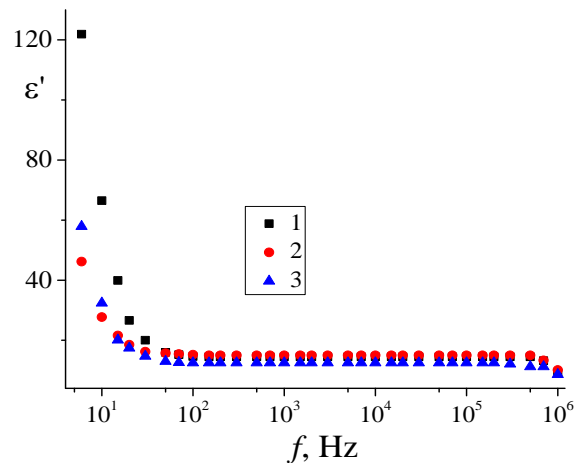


Fig. 1. Frequency dependences of the real component of the complex permittivity ε' for the homeotropically oriented nematic liquid crystal 6CB doped with the impurities SOR5 (1), SOR10 (2), and SOR15 (3). The impurity concentration is 0.1 mass %, the thickness of the samples is 50 μm . The temperature is 293 K (nematic phase of the liquid crystal).

more important for analysis. As we noted in [8], a uniform distribution of the electric field across the sample thickness is typical for this range of frequencies. For homeotropic alignment of molecules, the dielectric permittivity in this frequency range must be equal to the dielectric permittivity along the long axis of molecules. This is observed for the SOR5 and SOR10 impurities, respectively, curves 1 and 2 in Fig. 1.

In the case of the SOR15 impurity, slightly smaller values of ε' were obtained than those for the homeotropic orientation of 6CB molecules. The reason for this may be not the strictly homeotropic alignment of 6CB molecules, but a randomly tilted alignment of these molecules due to the impact of the impurity on the alignment quality of the electrode. Minor changes in the alignment capabilities of the electrode can be caused by the partial adsorption of impurity molecules on the electrodes. A significant difference in the value of ε' at $f < 100$ Hz for three various impurities, as will be shown below, is the main reason for the change in the dielectric spectrum depending on the impurity type.

In the case of dielectric spectroscopy method, the frequency dependences of the real ε' and imaginary ε'' components of the complex dielectric permittivity ε^* have been mainly analyzed. The analysis of the data obtained by us showed that to compare the influence of SOR5, SOR10 and SOR15 impurities on the electrical properties of 6CB, it is better to analyze not the frequency dependence of ε'' , but the frequency dependence of the alternating current (AC) electrical conductivity σ_{AC} . As is known [8–11], the values of ε'' and σ_{AC} are related by the relation:

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

where ε_0 is the electric constant.

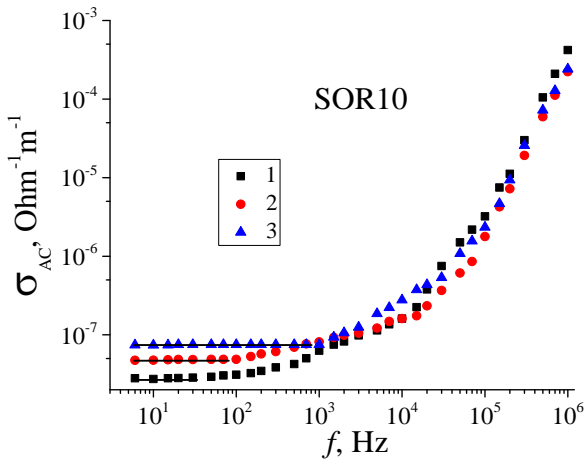


Fig. 2. Frequency dependences of the conductivity σ_{AC} of 6CB measured using three concentrations of SOR10 impurity: 0.01 (1), 0.05 (2), and 0.1 mass % (3). The sample thickness is 50 μm . The temperature is 293 K.

Fig. 2 shows the frequency dependences of the conductivity σ_{AC} of 6CB doped with the SOR10 impurity of three concentrations (0.01, 0.05 and 0.1 mass %).

From the analysis of the frequency dependences of σ_{AC} shown in Fig. 2, it is possible to draw an unequivocal conclusion – the highest difference in the conductivity depending on the concentration of the SOR10 impurity is observed at the frequencies $f < 10^3$ Hz. A characteristic feature of this frequency range is the presence of a frequency range where the value of σ_{AC} does not depend on the frequency f . In Fig. 2, these regions are marked with the solid lines. It is important to note that the width of this region (for the frequency range in which the research was carried out) is the largest at the maximum concentration of the SOR10 impurity (curve 3 in Fig. 2) and decreases with a decrease in the concentration of SOR10.

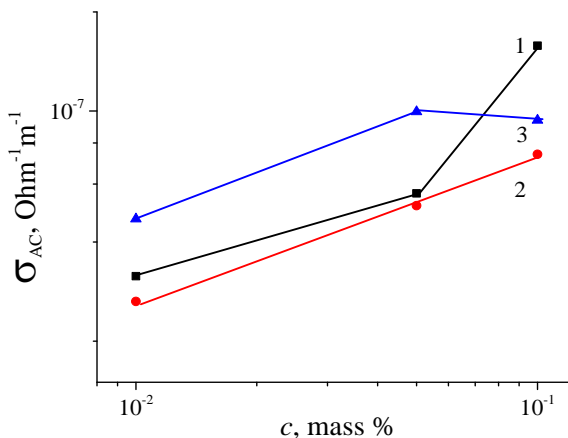


Fig. 3. Dependence of the ionic conductivity of the nematic liquid crystal 6CB on the concentration of impurities: SOR5 (1), SOR10 (2) and SOR15 (3). The sample thickness is 50 μm . The temperature is 293 K.

As is known [4, 5, 10, 11], the frequency regions where the value of σ_{AC} does not depend on the frequency correspond to the ionic conductivity of weak electrolytes, including LC. At the same time, it is important to know a type of quantitative dependence of the ionic conductivity of the liquid crystals (6CB) on the concentration of the introduced impurity. The dependence of the electrical conductivity of 6CB on the concentration of the SOR10 impurity is shown in Fig. 3 (curve 2).

From the analysis of the dependence of the ionic conductivity of 6CB on the concentration of SOR10 impurities (curve 2), it can be concluded that for a given range of concentrations it can be described by a power law, *i.e.*

$$\sigma_{AC} = ac^{0.48}, \quad (2)$$

where a is some constant. Taking the measurement error into account, the value of the exponent in the relation (2) is 0.48 ± 0.04 . That is, the power factor found from the experimental data for the dependence of the 6CB conductivity on the concentration of SOR10 impurities is close to 0.5. As is known [12], it corresponds to the bimolecular recombination of charge carriers. If we assume that nanoimpurities can be a source of ions [13–15], then similar behavior can be predicted for molecular liquid crystals with nanoparticles [16]. As was shown in the paper [17], even minor degree of ionic contamination of nanodopants can result in the behaviour similar to that of weak electrolytes in liquid crystals described by equation (2).

The complex behavior (Fig. 3) can be explained by taking into account several competing processes. Nanoimpurities are coated with oleic acid to prevent aggregation [1]. It is known that these nanoparticles can act as a source of ions, which leads to an increase in ionic conductivity [13–17]. This behavior is observed in its purest form for the SOR10 impurity (Fig. 3, curve 2). For a given level of ionic contamination of nanodopants and liquid crystals, there is a saturation effect, *i.e.*, the electrical conductivity reaches its saturation level and does not increase anymore [16, 17]. However, even in this case the ions can still be adsorbed on the electrode surface, leading to a decrease in electrical conductivity with an increase in the concentration of nanoimpurities (SOR15 impurities, Fig. 3, curve 3). Under some circumstances, the alignment layer can also act as a source of ions, increasing the ionic conductivity. For the same mass concentration, nanoparticles of smaller size have a higher volume concentration. This can lead to aggregation of nanoparticles and desorption of oleic acid with its subsequent dissociation. As a result, it will lead to a sharper increase in ionic conductivity (SOR5 impurities, Fig. 3, curve 1).

Fig. 4, as well as Fig. 2, shows the frequency dependence of the AC electrical conductivity of 6CB doped with several concentrations of the SOR5 impurity. The obtained dependences $\sigma_{AC}(f)$ (Fig. 4) differ a little from the data shown in Fig. 2. The main difference is that the frequency range, where the value of σ_{AC} does not depend on the frequency of the measuring signal f , is wider than in the case when the SOR10 impurity was added to 6CB.

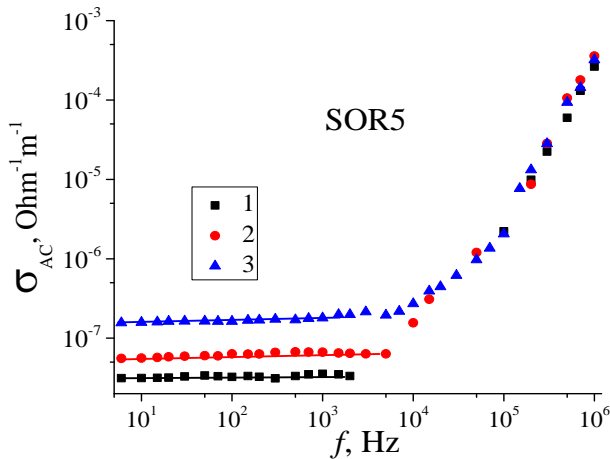


Fig. 4. Frequency dependences of conductivity σ_{AC} of 6CB measured using three concentrations of the SOR5 impurity: 0.01 (1), 0.05 (2) and 0.1 mass % (3). The sample thickness is 50 μm . The temperature is 293 K.

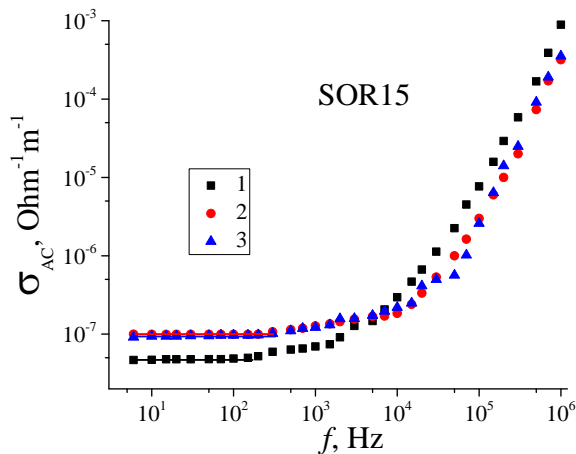


Fig. 5. Frequency dependences of conductivity σ_{AC} of 6CB measured using three concentrations of SOR15 impurity: 0.01 (1), 0.05 (2) and 0.1 mass % (3). The sample thickness is 50 μm . The temperature is 293 K.

Like in the case of the SOR10 impurity, the dependence of σ_{AC} value on the concentration of the SOR5 impurity was analyzed in more detail. This dependence is shown in Fig. 3 (curve 1). In contrast to the data obtained for the SOR10 impurity in Fig. 3 (curve 2), a power dependence of the conductivity on the impurity concentration is not observed for an entire range of concentrations. As was already mentioned, minor changes in physical-chemical composition of the nano-impurity (the degree of ionic contamination, the surface coverage by surfactants, *etc.*) can significantly affect the electrical properties of LC doped with such dopants.

Having observed a significant difference between the effect of SOR5 and SOR10 impurities on the electrical properties of LC 6CB, it was logical to check whether the effect of the SOR15 impurity would differ from that of the SOR5 and SOR10 impurities.

Fig. 5 shows the frequency dependence of the AC electrical conductivity of liquid crystals 6CB doped with the SOR15 impurity of varying concentrations.

The obtained frequency dependences of conductivity σ_{AC} of 6CB doped with SOR15 impurity at three concentrations shown in Fig. 5 differ a little from the data shown in Figs 2 and 4. The main difference is that, at the frequencies higher than 10^3 Hz, the conductivity of samples containing the lowest SOR15 impurity concentration of 0.01 mass % (curve 1) differs more significantly than in the case of SOR5 and SOR10 impurities from the data obtained for other concentrations. Because for all three impurities, the largest changes in conductivity occur precisely at the frequencies $f < 10^3$ Hz, we will not analyze the cause of this effect. The analysis of the concentration dependence of the ionic component of the LC conductivity is more important.

Like in the case of SOR5 and SOR10 impurities, the concentration dependences of σ_{AC} of 6CB containing three concentrations of the SOR15 impurity are shown in Fig. 3 (curve 3). From the analysis of the obtained data, it follows that these dependences are significantly different from those in the case of the presence of SOR5 or SOR10 impurities in 6CB. Similar to the case of SOR5 impurity, the concentration dependence of conductivity cannot be described by a power law over an entire concentration range. For concentrations higher than 0.05 mass %, further increase in the concentration of SOR15 impurity (from 0.05 up to 0.1 mass %) leads to a decrease in the value of σ_{AC} . At the same time, the maximum value of conductivity is observed for the impurity concentration of 0.05 mass %.

4. Conclusions

1. It has been shown that the SOR5, SOR10 and SOR15 impurities do not significantly affect the value of the real component of the complex dielectric permittivity (ϵ'). The largest changes are observed for the lowest frequencies ($f < 10^2$ Hz) and may be caused by the different influence of the impurity on the near electrode processes in liquid crystal samples.

2. For all types of impurities, the largest changes in the LC conductivity are observed in the frequency range $f < 10^3$ Hz. Because the value of the conductivity in this frequency range does not depend on frequency f , it corresponds to the ionic component of the conductivity through LC.

3. In the case of the SOR10 impurity, the conductivity of 6CB increases with an increase in the impurity concentration according to a power law with an exponent close to 0.5, which is typical for bimolecular recombination of charge carriers (similar to the behaviour of weak electrolytes in molecular liquid crystals).

4. For impurities SOR5 and SOR15, the concentration dependence of conductivity can be approximated by a power law only for concentrations below 0.05 mass %.

5. For the concentrations of SOR5 impurity higher than 0.05 mass %, the value of conductivity sharply increases. Under similar conditions, 6CB containing SOR15 impurity exhibits the opposite behavior. For the concentrations of SOR15 impurity higher than 0.05 mass %, the value of conductivity decreases, so that the maximum value of conductivity is observed at 0.05 mass % of SOR15 impurity.

6. The observed qualitative and quantitative changes in the electrical properties of nematic liquid crystals 6CB doped with SOR impurities can be associated with minor changes in chemical and physical composition of nanoimpurities.

Acknowledgements

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Authors' contributions

Garbovskiy Y.A.: conceptualization, methodology, validation, analysis, data curation, writing – original draft, writing – review & editing, visualization.

Kopčanský P.: resources, investigation, visualization, writing – review & editing.

Kovalchuk O.V.: formulation of the problem, analysis, investigation, writing – original draft, writing – review & editing, visualization.

Kovalchuk T.M.: writing – review & editing, project administration.

Volokh L.V.: investigation, resources, software.

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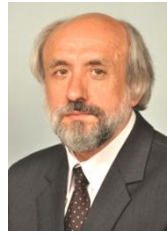
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Особливості впливу різного типу нанодомішок SOR на величину іонної складової електропровідності гомеотропно орієнтованого нематичного рідкого кристала 6CB

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Анотація. У цій статті повідомлено про вплив нанодомішок SOR5, SOR10 і SOR15 на величину іонної провідності гомеотропно орієнтованого нематичного рідкого кристала 6CB. Електричні вимірювання проведено в широкому діапазоні частот (від 6 Гц до 10^6 Гц) при кімнатній температурі (293 К). Показано, що найбільші зміни електропровідності відбуваються в області низьких частот (менше ніж 10^3 Гц) і залежать від типу нанодомішки та її концентрації. Незважаючи на подібність хімічного складу SOR5, SOR10 і SOR15, виміряні залежності електропровідності досліджуваних зразків від концентрації нанодомішок суттєво відрізняються. У випадку домішки SOR10 іонна складова електропровідності залежить від концентрації SOR10 за степеневим законом із показником приблизно 0,5, що характерно для бімолекулярної рекомбінації носіїв заряду (тобто поведінка як слабкий електроліт). Зразки рідких кристалів, що містять нанодомішки SOR5 і SOR15, поведуться подібним чином в області низьких концентрацій (між 0,01 і 0,05 мас. %). Цікаво, що подальше збільшення концентрації нанодомішок ($> 0,05$ мас. %) приводить до різко іншої поведінки. Електропровідність рідких кристалів з домішкою SOR5 різко зростає, тоді як електропровідність зразків, що містять SOR15, зменшується.

Ключові слова: діелектричні властивості, нематичний рідкий кристал, гомеотропне розташування молекул, іонна провідність, концентрація нанодомішок.