

Features of dielectric properties of medical thermal indicators based on dispersions of cholesteric liquid crystals in the polymer matrix

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Abstract. Within the frequency range $10 \dots 10^6$ Hz, the frequency dependences of the real (ϵ') and imaginary (ϵ'') components of the complex dielectric permittivity of medical thermal indicators based on polyvinyl acetate and mixtures of cholesteric liquid crystals have been studied. In them, the selective reflection of electromagnetic waves visible to the human eye occurs at normal (36.6 °C) and elevated (38.2 °C) human body temperatures. Being based on the comparison of the ϵ' frequency dependences for the studied in this work dispersions of nematic liquid crystals prepared using the same technology and with the same polymer, it has been shown that, already on the basis of analysis of frequency dependences for ϵ' , it is possible to ascertain the difference in characteristics for two types of thermal indicators. From the comparison of frequency dependences for ϵ'' , the main reasons of a difference between dielectric properties of the investigated medical thermal indicators for various temperatures of a human body have been ascertained.

Keywords: liquid crystal thermal indicator, selective light reflection, cholesteric liquid crystal, nematic liquid crystal, polymer matrix, dielectric spectroscopy.

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1. Introduction

Measuring the temperature, in particular the human body one, is an important technical task. In general, it has long been solved, but there are still many problems with implementation of simple rapid methods for measuring the temperature. The task of developing and implementing such methods has become especially relevant since the end of 2019 because of spreading the COVID-19 coronavirus epidemic. It is caused by the fact that currently the assessment of the surface temperature of the human body is one of the simplest and most common methods for identifying potential patients with COVID-19 in various types of institutions (including educational), organizations and transport. In principle, for this diagnostics it is not necessary to know exactly the temperature with an accuracy of 0.1 °C, but only to have

diagnostic methods that can indicate exceeding normal temperature of the human body by 1-2 °C.

Previously, mercury thermometers were widely used to diagnose the human body temperature. Because mercury vapors without introduction of certain, generally harmful for the human organism, substances themselves are not excreted from the organism and in high concentrations lead to severe disease, currently the use of mercury thermometers is practically prohibited. Therefore, it is important to search for other (than using mercury thermometers) simple methods for measuring the temperature.

Up to date, diagnostics of temperature in medicine is carried out mainly using electronic thermometers. But these thermometers are not cheap (and therefore not available for mass use) and in a portable version need batteries for their operation.

It has long been shown that cholesteric liquid crystals (CLC) or mixtures of nematic liquid crystals (NLC) containing “twisting” impurities can be used to measure the temperature [1-5]. From the physical point of view, it is related with the morphology of these crystals, namely: the arrangement of long axes of CLC molecules or in nematic-cholesteric mixtures in the form of spirals. In this case, the step of the spiral, the value of which increases with temperature, defines the wavelength for selective reflection of light. That is, by selecting a certain type of liquid crystal or mixtures of liquid crystals (which is mainly used in practice), one can create a thermal indicator, in which the selective reflection of electromagnetic radiation for the visible spectral range would occur at a specific temperature. Unlike electronic thermometers, batteries are not required for operation of these thermal indicators.

At the very beginning of implementation of CLC in the form of thermal indicators for medical purposes, they were directly applied to the surface of the part of human body, which temperature should be measured. This method of measuring temperature was inconvenient because, firstly, the layer of liquid crystal applied to the surface was difficult to reuse, and secondly, there was a problem with washing away the layer of CLC after measuring the temperature, since cholesterol molecules are fats and easily penetrate into pores of human skin.

The situation changed significantly, when thermal indicator films began to be used for measuring the temperature. In these films, the CLC or nematic-cholesteric mixture was dispersed into a polymer matrix. The most common method of dispersion is mixing (mechanical or ultrasonic) CLCs at a temperature above the melting point of the liquid crystal with a polymer solution or using the solvent in which both the polymer and liquid crystal are soluble. If the same solvent is used for CLC and polymer, it is not necessary to heat the solution of polymer and liquid crystal.

To obtain a film by using a special facility, a mixture of polymer and CLC is deposited onto horizontal surfaces with minimal adhesion of polymer to these surfaces. After drying, a layer of the required thickness is formed (in most cases hundreds of micrometers), in which due to separation of the phases after evaporation of the solvent, droplets of CLC are formed, which are surrounded by a polymer matrix. The diameter of the droplets is tens to hundreds of micrometers, which is much larger than the step of the cholesteric spiral (or the wavelength visible to the human eye range of electromagnetic waves). Therefore, even in these small droplets of CLC, the cholesteric spirals are not deformed and remain practically the same as those in thick films.

Since the thermal indicators based on the CLC “operate” using the effect of selective light reflection, to increase their efficiency in the case of using dispersions of these crystals in the polymer matrix, it is necessary to eliminate the effects of light reflection from the polymer (matrix) itself. To make it, some substance is added to the polymer solution before introduction of CLC. This

substance effectively absorbs electromagnetic waves for the entire visible range. Soot is the most available and cheapest substance of this type. Therefore, thermal indicators based on CLC or nematic-cholesteric mixtures must have at least 3 components: polymer, CLC (or preferably a mixture of CLCs) and carbon black.

For wider implementation of thermal indicators based on CLC, it is necessary to know their characteristics. From the analysis of published data [1-5], it follows that the study of the properties inherent to dispersions of CLC in the polymer matrix was mainly carried out at lower concentrations of CLC than those used in practice and in the absence of soot impurities.

The purpose of this work was to study the dielectric characteristics for two types of thermal indicators based on CLC dispersed in a polymer matrix with carbon black impurities, selective light reflection of which occurred at normal (36.5 °C) and elevated (38.2 °C) human body temperatures, and also to compare these results with our previously published data on the properties of NLC in the same polymer [6].

The main task of this study was to establish the possible difference in the electrical parameters of these films that differed only in different mixtures of NLC and were prepared on the basis of the same technology. This makes it possible to expand the methods of diagnostics of these thermal indicator films after their manufacturing. When comparing the results with data on the dielectric properties of dispersions for nematic liquid crystals in the polymer matrix, the main task was to identify and explain the possible reasons for the difference in dielectric properties of dispersions of liquid crystals in the polymer matrix when changing from NLC to the CLC mixture.

2. Materials and methods of measurement

To study, we used two types of CLC mixtures that were previously manufactured by the factory “Monocrystal Reagent” in Kharkiv.

For our research from the viewpoint of medical applications, it was important that the temperature range for selective light reflection in these mixtures corresponded to normal (36.5 °C) and elevated (38.2 °C) human body temperatures. These temperatures correspond to the green color, when being observed in the light reflected from the films, which is the most “receptive” to the human eye. The temperature range for the entire range of visible to the human eye part of the electromagnetic spectrum for the studied mixtures did not exceed 1 °C.

Technology of manufacturing dispersions of CLCs in the polymer was practically the same as in the work [6]. To prepare dispersions, we used a 10% aqueous solution of polyvinyl acetate. This solution was heated to the temperature close to 40 °C, and carbon black was introduced into it by mechanical stirring, followed by sonication. As in [7], we used N220 carbon black (Kremenchug Plant of Technical Carbon, Ukraine). The concentration of carbon black was 20% with respect to the polymer weight. Into the resulting mixture of an

aqueous solution of polymer with carbon black at the same temperature, we added a CLC mixture, in which the selective reflection of light (for green color) occurs at the temperature 36.5 °C (sample 1) or 38.2 °C (sample 2). The mass of CLC was the same as that of polymer with carbon black. Given the 5 times less mass of carbon black relative to the polymer, we can assume that the CLC mass is approximately equal to the mass of the polymer.

After embedding the CLC into the solution of polymer with carbon black, stirring and sonication were performed again. Mechanical stirring was carried out after turning off the heater and was continued until the sample was cooled to room temperature. At room temperature, by using a special facility a mixture of the aqueous solution of polymer, carbon black and CLC was deposited onto a glass plate. To avoid adhesion of the film (consisting of polymer with impurities of soot and CLC) formed after drying to the surface of the glass plate, before placing the solution onto these plate a thin layer of fat was used.

After evaporation of the solvent (water), the polymer film with CLC droplets dispersed in it was formed. The film thickness for two types of CLC mixtures (selective light reflection occurs at the temperatures 36.6 °C and 38.5 °C) was close to 150 μm.

Our studies by using an electron microscope at the voltage 20 kV (like that we used in [7]) showed that the diameter of the CLC droplets in the polymer matrix is close to 100 μm. Given that when observing in the visible to the human eye range of the spectrum of electromagnetic waves, the step of the cholesteric helix is close to 1 μm, the following can be stated: in the manufactured dispersions of CLC in the polymer matrix there were no obstacles to the “untwisting” of the cholesteric spiral in droplets, the size of which is much (more than 100 times) larger than the step of the cholesteric spiral. That is, we can assume that morphology of CLC in droplets did not differ significantly from morphology of CLC in the samples with the size much larger than 100 μm.

As in [7, 8], the dielectric properties of the obtained sandwich samples were investigated within the frequency range $6 \dots 10^6$ Hz by using the oscilloscopic method [9]. As electrodes, we used glass plates coated with a conductive layer of ITO (mixtures of tin and indium oxides). To ensure contact between the polymer film with carbon black and CLC droplets, thin layers of immersion oil were applied to the electrodes. The presence of electrodes transparent in the visible spectral range enabled to visually control the presence of selective light reflection due to illumination of the sample.

When measuring the dielectric properties of the samples, the amplitude of the measuring signal of the sinusoidal shape was 1.0 V. The amplitude of the measuring signal, which is higher than in [7], could be used due to the fact that electro-optical effects in CLC occur at electric fields higher by more than one order of magnitude than those used in NLC. Assuming that the equivalent circuit of the measuring cell is a resistor and

capacitor connected in parallel, we determined the values of resistance R and capacitance C of the samples at various frequencies, and being based on these values and geometric dimensions of the cell we calculated imaginary (ϵ'') and real (ϵ') components of the complex dielectric permittivity, respectively. The main results and conclusions of this work will be formulated exactly on the basis of the analysis of the reasons for differences in the frequency dependences of ϵ' and ϵ'' of dispersions of CLC in the polymer matrix for two types of samples, selective reflection of light in which occurs at the temperatures 36.6 °C and 38.5 °C, as well as comparing these data with the frequency dependences of the dispersions of NLC obtained in [7].

3. Results and their discussion

3.1. Frequency dependences of the real component ϵ' of the complex dielectric permittivity

Fig. 1 shows the frequency dependences of the real component ϵ' of the complex dielectric permittivity for the studied in this work sample 1 (dispersions of CLC in the polymer matrix, selective reflection of light at the temperature 36.6 °C) (curve 1), sample 2 (dispersions of CLC in the polymer matrix, selective reflection of light at the temperature 38.5 °C) (curve 2) and for the studied in [7] sample 3 dispersion of NLC in the polymer similar in composition (polyvinyl acetate), as CLC, at the temperature 293 K.

From the analysis of data shown in Fig. 1, the following conclusions can be drawn. Although the ϵ' values for the samples 1 and 2 differ from each other, their frequency dependences practically correlate with each other. Since the same polymer was used to prepare the samples 1 and 2, it is obvious that the difference in the ϵ' value of these samples is caused by the difference in

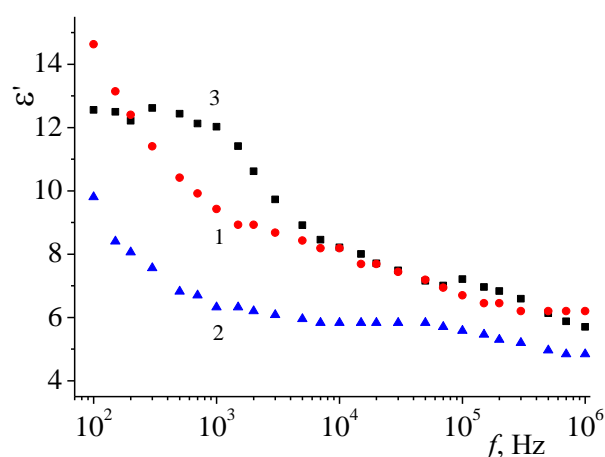


Fig. 1. Frequency dependences of the real component ϵ' of the complex dielectric permittivity for the dispersions of CLC in the polymer matrix sample 1 (selective reflection of light at the temperature 36.6 °C) (curve 1), sample 2 (selective reflection of light at the temperature 38.5 °C) (curve 2) and for the studied in [7] dispersion of NLC in the same polymers (polyvinyl acetate) as CLC at the temperature 293 K.

the dielectric properties of the CLC mixtures. As it is known [10], the main component of the dielectric permittivity of polar substances that include CLC is the dipole component. Therefore, the difference in the ϵ' values for the samples 1 and 2 can be explained by the fact that the dielectric permittivity of the NLC mixture used to prepare the sample 2 was higher than that for preparing the sample 1. As for the frequency dependences of NLC (curve 3), as it follows from Fig. 1, the frequency dependence is close to that of the sample 2. Given the fact that for the manufacture of all of the samples shown in Fig. 1, the same polymer was used, it can be assumed that the dielectric properties of the CLC mixtures used in the sample 2 are closer to the properties of NLC than to the dielectric properties of CLC mixtures used to prepare the sample 1.

In the solid state, the properties of liquid crystals differ a little from those of amorphous solids, including polymers (namely, in this state the CLC mixtures were shown in Fig. 1). Therefore, it was important how the ϵ' value will be different for samples 1 and 2 under the conditions when CLC transforms from the solid phase to the liquid crystal one. Accounting the narrow temperature range of existence of the cholesteric phase (below 1 °C as noted above), the comparison of these data was performed at different temperatures 36.6 °C and 38.5 °C. Since in [7] the dielectric properties of NLC dispersions at the temperatures close to the above were not studied, we will only analyze the difference between the dielectric properties of the samples 1 and 2 at the above temperatures.

As it follows from Fig. 2, the dielectric spectra of CLC mixtures dispersed in the polymer matrix (parameters of the samples 1 and 2) at the temperatures 36.6 °C and 38.5 °C differ much more than those at the temperatures 293 K (Fig. 1). The fact that at the temperatures 36.6 °C and 38.5 °C, respectively, the samples 1 and 2 were in the liquid state is confirmed by a more pronounced dispersion of the ϵ' value, depending on the frequency of the measuring signal, than that shown in Fig. 1.

As we noted in [6], the reason for this dispersion of the ϵ' value is the influence of near-electrode processes. Quite a significant difference between the dielectric properties of the samples 1 and 2 at the frequencies less than 1 kHz can be explained by a significant difference in the parameters of the near-electrode processes in these samples. As shown above, since the samples 1 and 2 differ by CLC mixtures different in compositions, it can be argued that in the case of studying the properties of dispersions of CLC in the polymer matrix under conditions when CLC transforms from the solid phase to liquid crystal one, the values of ϵ' and ϵ'' become more significantly dependent on the properties of the liquid crystal than in the case when CLC is in the solid state (Fig. 1). If to compare the dielectric spectra of ϵ' for the samples 1 and 2 with the data for the liquid phase of NLC, one can conclude that exactly the dielectric spectrum of the sample 2 (curve 2 in Fig. 2) is more similar to the dielectric spectrum of NLC. We believe

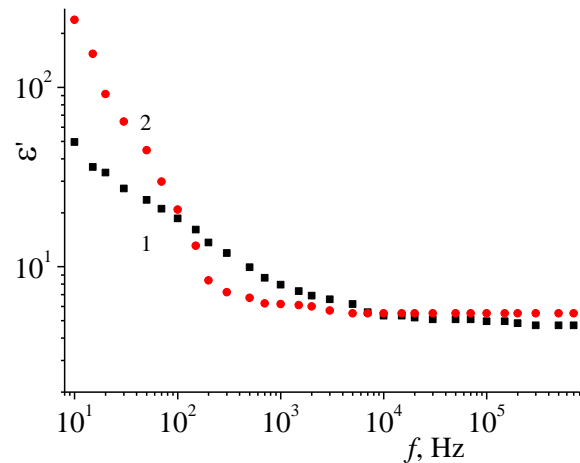


Fig. 2. Frequency dependences of the real component ϵ' of the complex dielectric permittivity of CLC dispersion in the polymer matrix for the sample 1 at the temperature 36.6 °C (curve 1) and the sample 2 at the temperature 38.5 °C (curve 2).

that the reason for this effect can be the difference in the distribution of liquid crystal droplets along the sample thickness found in the work [7].

As we showed in [7], when studying NLC dispersed in the polymer matrix and analyzing the microphotographs obtained using a scanning microscope, it is seen that in the polymer matrix after completion of the dispersion process due to evaporation of the solvent, areas can form (“holes” in the polymer), which are filled with liquid crystal over the entire sample thickness. Obviously, “holes” of similar types were formed when preparing the sample 2. In the case of the sample 1, these “holes” were either not formed, or their amount was much smaller than that in the sample 2.

3.2. Frequency dependences of the imaginary component ϵ'' of the complex dielectric permittivity

Fig. 3 shows the frequency dependences of the imaginary component ϵ'' of the complex dielectric permittivity for the sample 1 (curve 1), sample 2 (curve 2) and sample of the studied in [7] dispersion of NLC in the same polymer (polyvinyl acetate) as CLC (curve 3). All the measurements were carried out at the temperature 293 K.

If to compare the results shown in Fig. 3 with the data shown in Fig. 1, one can draw the following conclusions. First, like to for the frequency dependences of ϵ' , the frequency dependences of ϵ'' for the dispersions of NLC in the same polymer as CLC are closer to similar data for the sample 2. Second, by contrast to the frequency dependences of ϵ' (Fig. 1) the largest values of ϵ'' are observed exactly for the sample 2. As it follows from Fig. 1, at the same time the ϵ' values of the sample 2 were smaller than those for the sample 1 (curve 1) and for NLC dispersions (curve 3).

As it is known [10], the ϵ' value characterizes the changes related with polarization effects in substance, and the ϵ'' value – with dielectric losses. For the studied frequency range, dielectric losses are mainly caused by

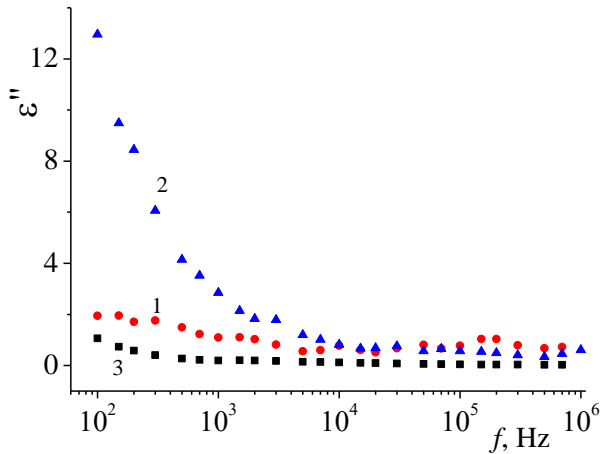


Fig. 3. Frequency dependences of the imaginary component ϵ'' of the complex dielectric permittivity for the sample 1 (selective reflection of light at the temperature 36.6 °C) – curve 1, sample 2 (selective reflection of light at the temperature 38.2 °C) – curve 2, and the sample of the studied in [7] dispersion of NLC is in the same polymers (polyvinyl acetate) as CLC – curve 3. All the measurements were carried out at the temperature 293 K.

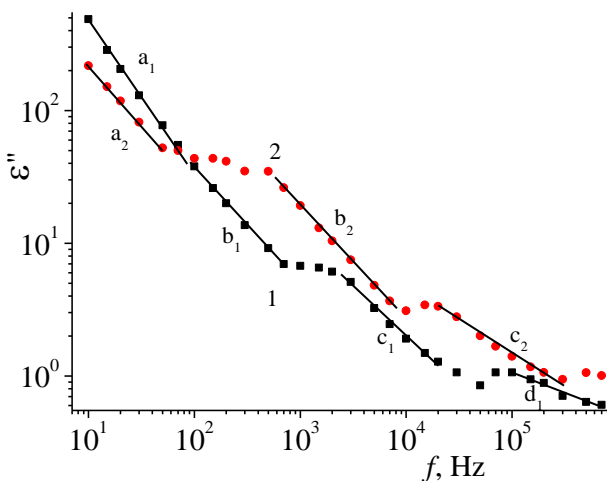


Fig. 4. Frequency dependences of the imaginary component ϵ'' of the complex dielectric permittivity for the sample 1 – curve 1 and sample 2 – curve 2. Solid lines marked areas (a, b, c, d) of power dependence of the ϵ'' value on the frequency.

conduction currents. Therefore, from the above data we can conclude that the sample 1 in relation to the sample 2 is characterized by better dielectric properties and lower dielectric losses.

As noted above, at room temperature, the mixtures studied in this work were in the solid state. Therefore, it was logical, as in the case of the analysis of frequency dependences of ϵ' , to consider the frequency dependences of ϵ'' for temperatures corresponding to the selective reflection of light. These dependences are shown in Fig. 4.

From the analysis of the data shown in this figure, it follows that for almost the entire frequency range (except for frequencies less than 100 Hz) the ϵ'' value for the sample 2 exceeds the ϵ'' value for the sample 1. From our viewpoint, this difference is caused by the fact that the dielectric properties of samples at low frequencies characterize the near-electrode processes, and at higher frequencies – the bulk properties of the substance. Therefore, from the obtained data it can be concluded that the CLC mixture introduced into the polymer matrix for the sample 1 had a more pronounced effect on the near-electrode processes, and the CLC mixture for the sample 2 had a stronger effect on the bulk properties of polymer.

As it is known [10], the ϵ'' value is related with the electrical conductivity of substances for alternating current σ_{AC} in accord with the relation

$$\sigma_{AC} = \epsilon'' \epsilon_0 \omega, \tag{1}$$

where ϵ_0 is the electric constant, $\omega = 2\pi f$ – cyclic frequency.

The power dependence of the ϵ'' value on the frequency of the measuring signal means that the σ_{AC} value in the areas of the dielectric spectrum marked by lines also changes with the frequency of the measuring signal according to the power law. That is, the relation is fulfilled, which in most monographs and articles is expressed not by frequency as a value inverse to the period, but by cyclic frequency to the period

$$\sigma_{AC} = a \omega^n, \tag{2}$$

where a is a constant that depends on the process of charge transfer for a particular area of frequency dependence, where the relation (2) is fulfilled, and n is the exponent for the frequency dependence of conductivity.

As it is known [11], the relation (2) describes the transfer of charge carriers (in most cases electrons) in solid disordered systems. For the samples studied in this work, polymer should be related to such systems. In the very liquid crystals, the conductivity does not depend on frequency. Thereof, one can conclude that for dispersions of CLC in the polymer, like to the dispersions of NLC in polymers studied in [7], the main limitation of electrical conductivity occurs in those parts of the sample where the polymer is available.

Among the parameters included to the relation (2), the most informative about the mechanism of charge transfer is the exponent n . Therefore, on the basis of the data shown in Fig. 4, its value was estimated for each area where the ϵ'' value and, respectively, the σ_{AC} value depended on the frequency in accord with the power law. The values of the exponents for each marked in Fig. 4 area of frequency dependence of the imaginary component of the complex dielectric permittivity for the samples 1 and 2 are listed in Tables 1 and 2, respectively.

Table 1. Values of exponents n in different areas of the frequency dependence of ε'' for the sample 1.

Area	a_1	b_1	c_1	d_1
n	-0.18 ± 0.02	0.12 ± 0.02	0.25 ± 0.02	0.70 ± 0.02

Table 2. Values of exponents n in different parts of the frequency dependence of ε'' for the sample 2.

Area	a_2	b_2	c_2
n	0.11 ± 0.02	0.13 ± 0.02	0.50 ± 0.02

From the analysis of the data adduced in the tables, one can conclude that the exponent n for the frequency dependence of the conductivity increases with frequency. It is important to note that for the sample 1 at the lowest of the studied frequencies, it takes even a negative value. The reason for this effect may be a significant contribution of near-electrode processes to changes in the frequency dependence.

4. Conclusions

The above analysis of the experimental results obtained in this work can be summarized as follows.

1. The difference between the parameters of medical thermal indicators made on the basis of the same polymer and different compositions of mixtures of cholesteric liquid crystals can be ascertained with account of frequency dependences typical to the real component ε' of the complex dielectric permittivity. It has been shown that for studies at room temperature the frequency dependence of ε' can be close (sample 1) or different (sample 2) from the similar dependence for a nematic liquid crystal dispersed in the same matrix as for the CLC mixture. It has been found that being based on the analysis of frequency dependences of ε' , namely, the correlation of their type with the frequency dependences of homogeneous CLC, it is possible to estimate the presence of areas in such matrices that are filled with CLC over the entire film ("holes" in polymer).

2. The difference between the parameters of medical thermal indicators manufactured on the basis of the same polymer and mixtures of cholesteric liquid crystals different in compositions can also be ascertained using the analysis of frequency dependences for the imaginary component ε'' of the complex dielectric permittivity. It has been shown that, like to the case of the frequency dependence of ε' , the frequency dependence of ε'' obtained at room temperature may be close (sample 2) or significantly different (sample 1) from the frequency dependence of ε'' of the nematic liquid crystal dispersed in the same polymer matrix. The most significant difference between the frequency dependences of ε'' for two types of the samples is observed at the temperatures when selective reflection of visible to the human eye electromagnetic waves takes place. It has been assumed that this difference is caused by different compositions of impurities in liquid crystals, each of which manifests itself in another manner for different parts of the frequency dependence of ε'' .

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Особливості діелектричних властивостей термоіндикаторів медичного призначення на основі дисперсій холестеричних рідких кристалів у полімерній матриці

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Анотація. У діапазоні частот $10-10^6$ Гц досліджено частотні залежності дійсної (ϵ') та уявної (ϵ'') компонент комплексної діелектричної проникності медичних термоіндикаторів, виготовлених на основі полівінілацетату та сумішей холестеричних рідких кристалів. У них селективне відбивання видимих людським оком електромагнітних хвиль відбувається при нормальній ($36,6^\circ\text{C}$) та підвищеній ($38,2^\circ\text{C}$) температурах людського тіла. На основі порівняння частотних залежностей ϵ' для досліджених у даній роботі дисперсій нематичних рідких кристалів, виготовлених за такою же технологією і з таким же полімером, показано, що вже на основі аналізу частотних залежностей ϵ' можна встановити відмінність у характеристиках двох типів термоіндикаторів. З порівняння частотних залежностей ϵ'' встановлено основні причини різниці діелектричних властивостей досліджуваних медичних термоіндикаторів для різних температур людського тіла.

Ключові слова: рідкокристалічний термоіндикатор, селективне відбивання світла, холестеричний рідкий кристал, нематичний рідкий кристал, полімерна матриця, діелектрична спектроскопія.