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ELECTRICAL PROPERTIES OF SOME FATTY ACIDS - CHOLESTEROL MIXTURES

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Abstract Some electric properties of some fatty acids (arachidic, arachidonic, elaidic and lauric) and fatty acid-cholesterol mixtures were determined. The $I=I(U)$ plots revealed a hysteresis behaviour, the hysteresis area being dependent on the cholesterol content. From the $I = I(t)$ plots, the relaxation time τ and the space charge Q_{sp} were determined and it was shown that in case of arachidic acid-cholesterol mixtures 1:1 molar, τ and Q_{sp} reached maxima. The results are explained by considering a hopping mechanism for conduction.

Keywords: fatty acids, arachidonic acid, cholesterol, biological membranes

INTRODUCTION

Fatty acids are a class of biologically important lipids. They are constituents of membranes and intracellular organelles and play a role in some biological processes such as transmission of information across membranes.

Singer and Nicholson¹ have elaborated the mosaic model of the biological membrane. Later it has been shown that the fluidity of the membrane depends not only on the intrinsic or extrinsic proteins but also on the concentration of ions crossing the membrane or on the pH^{2,3,4}.

In some biological membranes, a liquid crystalline state was also found⁵.

The investigations concerning the mesomorphic behaviour of the various lipidic systems may be of importance

in elucidating some properties of the biological membranes connected to their fluidities or to their electrical behaviour.

This is the reason why we investigate, in this paper, several properties of some fatty acids, constituents or forerunner constituents of biological membranes, as well as the behaviour of mixtures of these fatty acids with cholesterol.

EXPERIMENTAL DATA

The following fatty acids were investigated : lauric acid ($\text{CH}_3(\text{CH}_2)_{10}\text{COOH}$), arachidic acid ($\text{CH}_3(\text{CH}_2)_{18}\text{COOH}$), elaidic acid ($\text{CH}_3(\text{CH}_2)_7\text{CH}=\text{CH}(\text{CH}_2)_7\text{COOH}$) and arachidonic acid. The investigated fatty acids exhibit thermotropic, enantiotropic mesomorphism of the smectic C-type within the following temperature ranges : $73-75.3^\circ\text{C}$ (arachidic acid), $35-45^\circ\text{C}$ (lauric acid), $40-51^\circ\text{C}$ (elaidic acid) and $3-19^\circ\text{C}$ (arachidonic acid)⁶.

Mixtures of these fatty acids with cholesterol: 2+1, 1+1 and 1+2 (molar) were also studied. The mixtures were obtained by solving the fatty acids in carbon tetrachloride in the desired percentage. After stirring and letting the solvent evaporate, liquid crystal cells 20 μm thick with transparent and conductive electrodes were filled by capillarity with the mixtures. All the investigated mixtures exhibited smectic C phase, the mesomorphic range being larger when compared to the pure fatty acids^{6,7,8}.

The mesogenic interval was enhanced when the fatty acids were subjected to high electric field strengths (10^4-10^5 V/cm) and when cholesterol or other additives were introduced into the mixtures^{6,8}.

Here we report some of the electrical measurements we made :

I = I(U) plots

The temperature was kept constant during the experiments.

the precision was $\pm 0.01^{\circ}\text{C}$; it was measured with the aid of differential copper-constantan thermocouples attached to the liquid crystal cell. At a desired temperature, the current was determined when the voltage was first increased and then decreased. The $I = I(U)$ plots revealed an electric hysteresis, which was dependent on the cholesterol percentage. In some investigated samples a domain of negative resistances was noticed when the voltage was increased

The $I = I(U)$ plot obtained for pure arachidonic acid at 9°C , within the smectic phase, is shown in Figure 1.

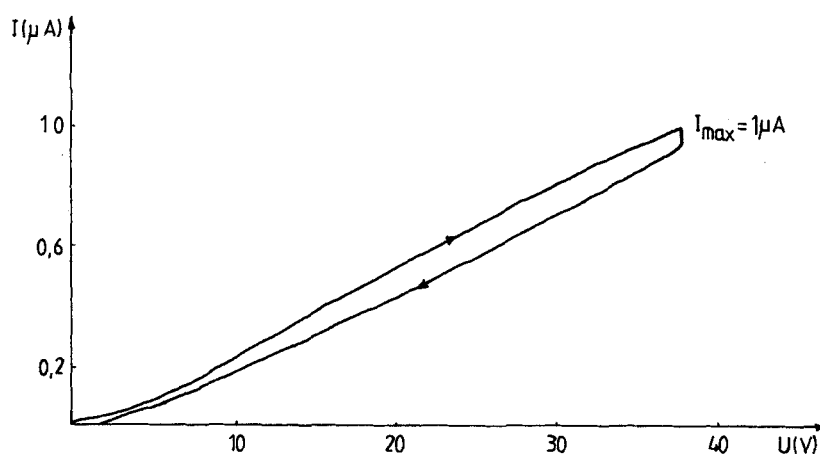


FIGURE 1 The $I=I(U)$ plot for arachidonic acid.

The hysteresis area is first increased when cholesterol is added to arachidonic acid (Figure 2) and then it is decreased when the cholesterol percentages are higher.

The conductivity of the mixture 1+2 is increased while the conductivity of the mixture 2+1 is decreased when compared to the one of the pure acid.

If a saturated acid (arachidic) or unsaturated acid (elaidic) is added to the mixture arachidonic acid-cholesterol 2+1, the conductivity is increased by one order of magnitude and a significant remanent polarization is noticed.

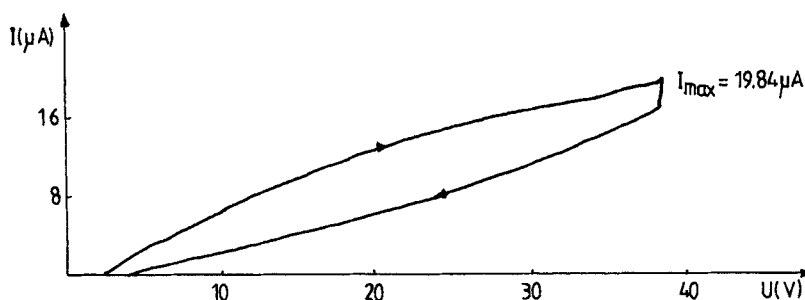


FIGURE 2 The $I = I(U)$ plot for the arachidonic acid-cholesterol mixture 1+2, obtained at 20°C (smectic C phase).

$I = I(t)$ plots

The temperature and the voltage were kept constant ; when the current reached a saturation value, the voltage was switched off and the evolution of the current was followed in time. In most cases the current is cancelled after a few minutes (Figure 3) ; in cholesterol-fatty acid mixtures 1+1 molar, no cancellation of the current was noticed, the sample remaining polarized several days (Figure 4).

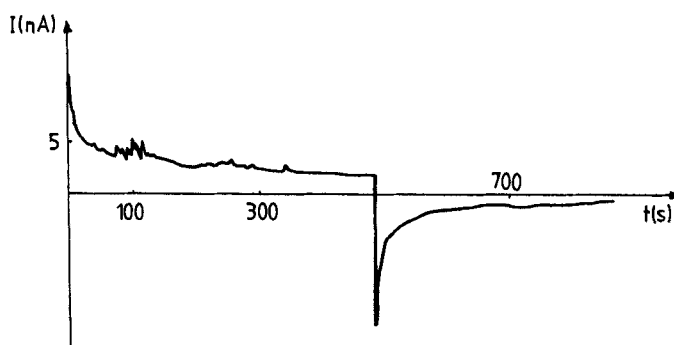


FIGURE 3 The $I = I(t)$ plot obtained at 20°C for arachidonic acid-cholesterol mixture 2+1 molar (smectic C phase).

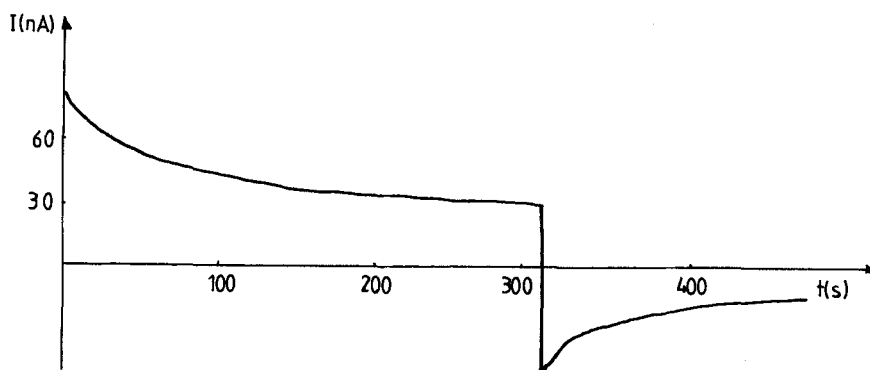


FIGURE 4 The $I = I(t)$ plot obtained at 20°C for arachidonic acid-cholesterol mixture 1+1 molar (smectic C phase).

This behaviour is similar to the one encountered in dielectrics exhibiting spontaneous polarization or in solids where the currents are space-charge limited.

The $I = I(t)$ plots allow for the determination of the relaxation time of charge carriers and of the space charge within the sample.

Using the method described by Tanev⁹, we obtained the results which are summed up in Table I.

DISCUSSION

It has been proved that, by adding cholesterol to fatty acids, spectacular changes occur in relaxation time of charge carriers, space charge accumulated within the sample and hysteresis area as determined from the $I = I(U)$ plots.

When mixtures of cholesterol with arachidic or arachidonic acid are examined, the effects are maximal in case of 1+1 percentages. When the cholesterol percentage is increased, as for instance in case of 1+2 mixtures, the conductivity is increased and the relaxation time is decreased when compared to the other cases.

TABLE I Relaxation time τ and space charge Q_{sp} for some fatty acids and mixtures⁺).

Substance	τ (s)	Q_{sp} ($\times 10^9$ C)
arachidic acid	125	10
lauric acid	100	8
arachidonic acid	29	1240
arachidonic acid- cholesterol mix- ture 2+1 (molar)	25	63.8
arachidonic acid- cholesterol mix- ture 1+1 (molar)	40	852
arachidonic acid- cholesterol mix- ture 1+2 (molar)	6	463

⁺) The results were obtained at temperatures within the smectic C range : 74°C (arachidic acid), 40°C (lauric acid), 9°C (arachidonic acid) 20°C (arachidonic acid-cholesterol mixtures).

This behaviour suggests that under such circumstances an additional phase is formed ; the mixture contains clusters of pure fatty acid and cholesterol embedded in the homogeneous mixture ; consequently, the conductivity of the mixture will be changed. The existence of these clusters was evidenced by microscopical investigations.

Such a case is known to occur when the interaction of water molecules with phospholipid molecules is considered in the presence of cholesterol¹⁰. The peculiar behaviour of such systems is explained by considering the effect of cholesterol on the polar groups of the phospholipids. The properties of this "bound water" are considerably different from those of the "bulk water".

As the systems investigated by us are not hydrated, it may be suggested that the effect of cholesterol on arachidic or arachidonic acid consists in changing the

space charge distribution ; in this way the mobilities of the ions or electrons will be changed when an external field is switched on.

The existence of this internal space charge will give rise to changes in the orientation of the molecules and their associated textures , as it was shown by us in other papers^{6,11}.

In order to explain the conduction mechanism in mixtures containing cholesterol, a theoretical model was proposed by one of us¹². This model is based on the following hypotheses :

1. When a d.c. electric field is switched on, each individual charge Δq attached to a molecule or to a cluster is shifted along virtual channels generated by the electric field and favoured by the presence of the cholesterol. This explains the increase of the conduction in mixtures containing high cholesterol percentages. The internal current will be obtained by a summation of the individual currents corresponding to each moving charge. The drift velocity of charges determines the dependence of the current on time. This model is similar to the one introduced by Langer et al¹³.

2. When the voltage is switched off, the charges attached to molecules or clusters are in excited states. In order to reach the equilibrium state, the system undergoes transitions to states of lower energies :

$$E_1 \rightarrow E_2 \rightarrow \dots \rightarrow E_{n+1} = A_0$$

where A_0 is the equilibrium energy level (ground state).

The electric charge is subjected to the internal field of the space charge, whose distribution was generated by the previous external field. The shifting of these charges through the virtual channels is described by a hopping mechanism characterized by a distribution of potential barriers.

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