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PHASE BEHAVIOUR OF A TERNARY SYSTEM CONTAINING LECITHIN

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Abstract The phase diagram of the ternary system egg yolk lecithin - water - hexadecanphosphondiethylesteric acid (PAE_{16}) is obtained. The phases were identified by means of phosphorus NMR and optical observations. Two regions of lamellar phases, one reverse hexagonal and the respective two-phase regions were found. The electroconductivity of all samples was measured. The correlation between the change of the conductivity and the phase transition is discussed.

EXPERIMENTAL METHODS

The phase diagram of the ternary system egg yolk lecithin (EYL) - water (D_2O) - hexadecanphosphonaciddiethylester (PAE_{16}) was studied.

This system was investigated by means of phosphorus NMR, optical observations and electroconductivity measurements.

The NMR investigations were carried out by means of a spectrometer HX 90 in a regime of spin decoupling. The identification of the liquid crystal phases was performed through the shape character of the absorption spectrum.

The optical observations were conducted by a microscope with cross polarizers equipped with thermostat table.

The electroconductivity was determined by means of conductometer OK 102/1 type provided with sample thermostating device.

The optical observations and conductivity measurements were performed in the temperature range of $15^{o}C$ to $55^{o}C$ in steps of $5^{o}C$. The NMR investigations proceeded at room temperature (about $25^{o}C$). The investigations of the ternary system were directed to the region of low water content of the phase diagram (below 50 % D_2O by weight).

RESULTS AND DISCUSSION

The phase diagram of the investigated ternary system was found (Fig.1) to be with two different regions of a lamellar phase $L_{\alpha A}$ and $L_{\alpha B}$, one hexagonal H as well as multiphase regions. The lamellar phase in the region of higher concentrations of egg yolk lecithin is well known from numerous previous investigations[1,2]. The transition from this lamellar phase to the hexagonal one when the admixture PAE_{16} is added, is similar to that previously observed by us[3] in the ternary system egg yolk lecithin (EYL) - water (D_2O) - alkanphosphonaciddiethylester (PAE_6). The transition from the above mentioned lamellar phase to the hexagonal one was observed through microscopic observations and electroconductivity investigations without addition of more admixture, only at temperature increase. Similar phase transition was reported by Marsh and Seddon[4] for other systems. The region of the hexagonal phase is considerably larger than in the above mentioned ternary system[3]. In the phase diagram of the system (EYL - D_2O - PAE_{16}) another region of lamellar phase appears at higher concentrations of

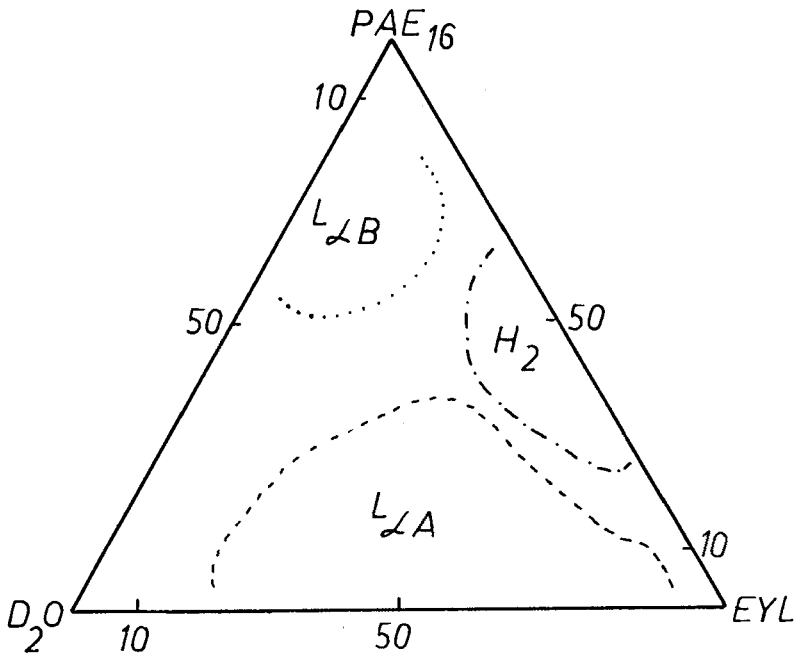

FIGURE 1. The phase diagram of (EYL) -(D_2O) - (PAE_{16}) obtained by NMR at 25°C

PAE_{16}. This behaviour of the ternary mixture is probably due to the well pronounced mesogenic character of PAE_{16}. As our observations show, the binary system (PAE_{16} - D_2O) forms a liquid crystal lamellar phase.

The electroconductivity of all samples was measured in the indicated temperature range.

Let us discuss the results from electroconductivity measurements of samples with different concentrations and changes in the temperature.

At first to compare the values of the elecroconductivity of three samples A (67% EYL, 17 % PAE_{16}, 16 % D_2O), B (20 % EYL, 58 % PAE_{16}, 22 % D_2O) and C (48 % EYL,

43 % PAE_{16}, 9 % D_2O) with different concentration composition at the same fixed temperature $t = 25^{o}C$ (at which the NMR measurements were made). The electroconductivity is relatively high for both lamellar phases A and B σ_A = 100 μS and σ_B = 130 μS respectively. In them the water layers, in which transport of ions is realized, are separated by the hydrophobic cores of the lamellas. The electroconductivity of the volume sample is due to a certain extent to the presence of structure defects, for instance hydrophilic passages through hydrophobic cores. Lowest electroconductivity was measured in the case of sample C (hexagonal phase) σ_C = 9.7 μS. In this phase, a continuous hydrophobic medium exists. The hydrated regions with high ion conductivity are almost entirely separated.

Let us consider the electroconductivity dependence as a temperature function.

On Figure 2A the temperature dependence of electroconductivity is plotted for sample A. Optical observations show that the sample preserves its nature in the whole temperature range (lamellar phase). In this case the electroconductivity increases monotonously with the rise in temperature owing to temperature activation of the mobility of carriers.

On Figure 2C is plotted the temperature behaviour of the electroconductivity of sample C. A monotonous increase in the electroconductivity similar to that with sample A is observed on increasing the temperature. Microscopic observations show that the hexagonal phase persists in the whole temperature range.

The character of the curve shown on Figure 2D, sample D (30 % EYL, 42 % PEA_{16}, 27 % D_2O) is quite different. At low temperatures, the ascending part of the curve corresponds to

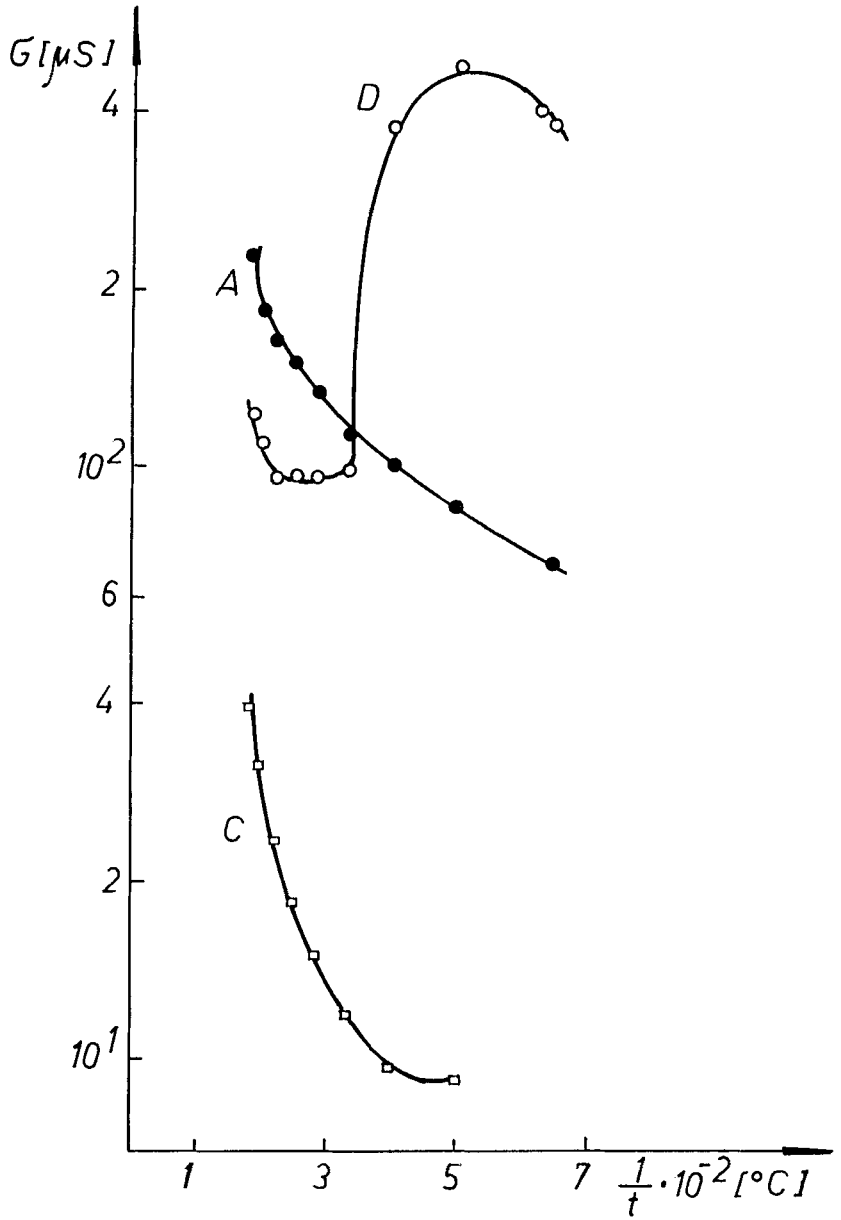

FIGURE 2. The temperature dependence of electroconductivity.

a lamellar phase (optically established as well). In the region of intermediate temperatures, a maximum and a minimum of the curve appear. We interpret this as a phase transition to a hexagonal phase. At higher temperatures, the hexagonal phase has already been established. The electroconductivity increases again which corresponds to temperature - activated mobility of the carriers but this time under conditions of lower conductivity.

CONCLUSION

We consider that the three methods used, namely, NMR, optical observations and electroconductivity measurements complement each other very well.

The NMR investigations played a major role in determining the phase diagram. The absorption NMR spectrum definitely show the phases in the system - lamellar, hexagonal, isotropic or multiphase.

The microscopic observations have an auxiliary role in determining the phases but they enable to define the presence or absence of phase transition in a wide temperature range in a comparatively simple and rapid manner.

The electroconductivity measurements provide convincing information about whether the structure contains water or hydrophobic continuum, which allows to distinguish the normal and the reversed hexagonal phases. The low electroconductivity of sample C warrants the assumption that the hexagonal phase obtained in our system is a reversed one.

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