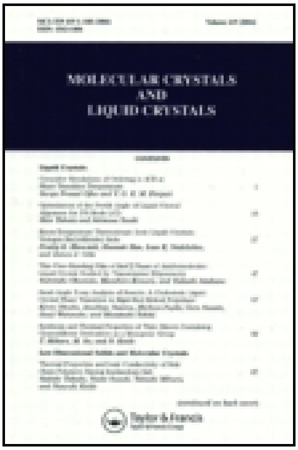

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DETECTION OF A NEW LIQUID CRYSTAL PHASE IN A BINARY SYSTEM BY THERMAL PHASE STUDIES

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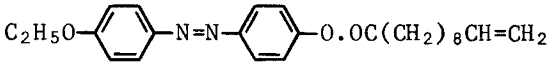
Abstract: Studies on the binary mixture EPAP undecylenate-tetrabutyl tin have been carried out at higher concentrations than previously. The mixture, during the cooling cycle, presents a new texture resembling 'Isogyres'. The photographs show the formation of the Isogyres along with the other phases. Preliminary comparisons of the Isogyric structure suggest that the new phase is cholesteric.

Introduction: It is well known that the order of mesophase stability on a scale of increasing temperature is Solid → Smectic → Nematic/Cholesteric → Isotropic. Though this order is generally true, recent investigations of binary mixtures of mesogenic cyano compounds by Cladis[1] and of pure compounds by Madhu Sudana *et al.*[2] have revealed the existence of reentrant nematic phase transitions on cooling. At elevated pressures, reentrant nematic phase transitions in pure cyano compounds have also been observed.[3,4] Such types of deviation from the general behaviour are relatively few in literature reports.

In the present work, a new deviation is reported for the first time for a binary mixture of a mesomorphic and a non-mesomorphic system.

EPAP undecylenate is the chosen nematic solvent and tetra-butyl tin, a non-correlating and non-mesomorphic solute, is



taken for investigation as the solute. Studies of phase diagrams of this binary mixture for lower concentrations have been reported elsewhere.[5] The present studies have been extended to higher solute mole fractions with a view to finding the composition range of nematic stability.

Experimental: The study of the phase transitions has been carried out using a polarizing microscope with a hot-stage. The accuracy in the measurement of temperatures is $\pm 0.25^{o}C$. Solute mole fractions from 0.30 onwards have been studied.

Previous studies[5] of the same binary system showed the presence of a two phase region for both heating and cooling cycles up to a solute mole fraction of 0.30. However, at a mole fraction of 0.319, a two-phase region is observed only while heating, and this phenomenon has continued up to 0.478 mole fraction. It is further observed that for concentrations higher than 0.478, the transition is from the solid to the isotropic phase directly. However, in the cooling cycle, an interesting new phase is observed at concentrations between 0.319 and 0.610.

Various stages of the process have been photographed with the sample between crossed polariser and analyser and using transmitted light.

Figure 1 shows the solid state of the binary mixture. Figure 2 presents the Nematic-Isotropic two-phase region, at a mole fraction of 0.380, in the heating cycle.

The interesting observation during the cooling cycle from the isotropic state for all the concentrations under study is the onset of a texture resembling 'Isogyres'[6] as depicted in Figure 3 for the concentration of 0.590. On further cooling, parallelogram-shaped flakes started appearing as in Figure 4. The flakes are initially seen as rods viewed along the plane of the parallelogram. In the next stage, these rods, rotating about themselves, gave rise to clear parallelograms as can be seen from Figure 5. The disappearance of Isogyres and the complete appearance of flakes in the entire field of view is displayed in Figure 6.

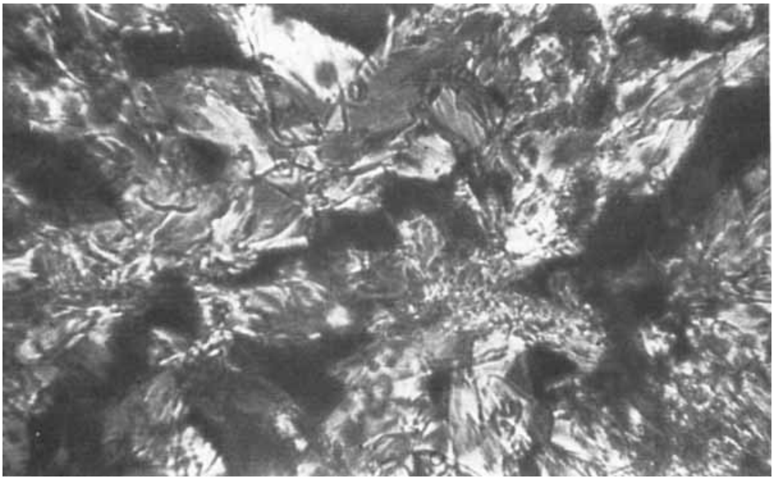


FIGURE 1 Solid phase

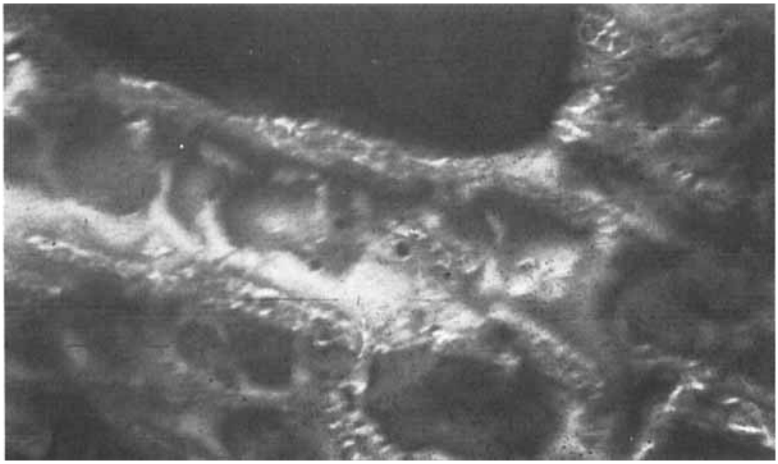


FIGURE 2 N-I two-phase region in the heating cycle

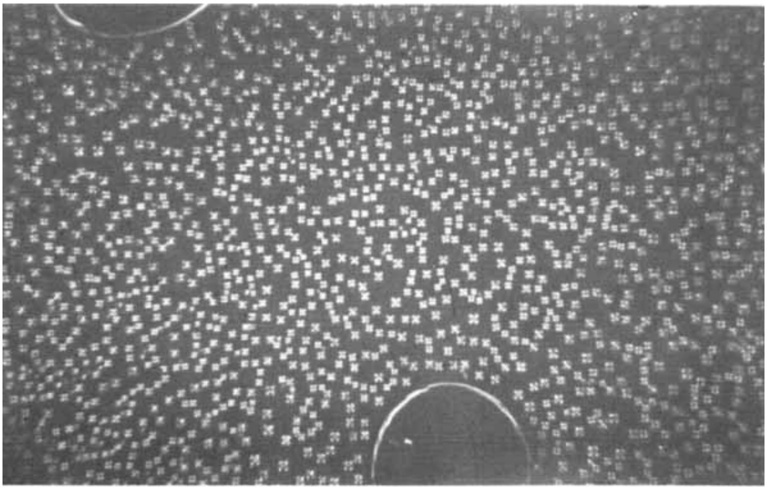


FIGURE 3 On-set of Isogyres at 0.590 concentration (cooling cycle)

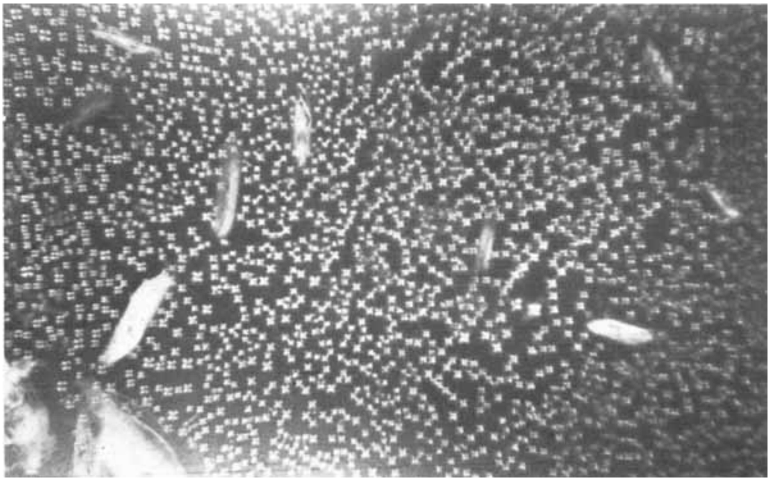


FIGURE 4 Formation of parallelogram-shaped flakes

FIGURE 5 Parallelogram texture after rotation of the rods

FIGURE 6 Texture near to the solid phase (cooling)

Discussion: Figures 1 and 2 bring out the usual features of the binary system. The occurrence of Isogyres in the cooling cycle (Figure 3) indicates a new phase transition, and comparison of the present textures with those found in the literature[6,7] suggest that this new phase is cholesteric. This is a surprising result, as the system is apparently non-chiral, but detailed and repeated investigation of the texture obtained on cooling precludes the possibility of any special texture of the nematic phase in this binary system.

For concentrations higher than 0.478, the Isogyric texture is retained in the monotropic state. A normal, reversible solid to isotropic transition is observed in both heating and cooling cycles for concentrations of 0.61 onwards. The various phases observed are presented in Table 1.

TABLE 1

Phases observed for various solute mole fractions of tetrabutyl tin

Solute mole fraction	Phase observed on Heating	Phase observed on Cooling	Type of transition
0.300	Nem → Iso	Iso + Nem	Enantiotropic
0.300 - 0.478	Nem → Iso*	Iso → Cholesteric*	Enantiotropic
0.478 - 0.610	Solid → Iso	Iso → Cholesteric	Monotropic
0.610 onwards	Solid → Iso	Iso → Solid	-

* The N-I and I-Ch temperatures are the same within 0.2°

The interesting difference in texture between Figure 1 and Figure 6 suggests the probable existence of another phase transition (possibly to a smectic phase) between the cholesteric and solid phases. This is because the parallelogram shaped flakes exist over a detectable temperature range (4°).

Further studies of the existence of these different phases using other techniques is under progress.

We are grateful to Professor GW Gray for all his kind help and encouragement.

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