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8 June 1979

Changed Lyotropic Liquid Crystalline Structure Due to Polymerization of the Amphiphilic Component

Abstract. *Optical patterns in polarized light and x-ray reflections in the low-angle region were used to detect a shift from one liquid crystalline structure to another during polymerization. The polymerization took place in a lyotropic liquid crystal of water and sodium undecenoate, with a structure consisting of cylinders in a two-dimensional hexagonal close packing. After polymerization, a lamellar liquid crystalline structure was obtained.*

The comprehensive knowledge of liquid crystalline structures (*1–4*) that has been obtained through their use for display systems and the excellent properties of Kevlar fibers drawn from liquid crystals (*5*) have resulted in a recent focus on polymerization in systems of liquid crystalline character. The contributions so far have mainly been in the area of thermotropic liquid crystals (*6*); the equilibrium problems being more complex in lyotropic structures, which by definition are multicomponent systems.

Early attempts to polymerize in lyotropic liquid crystals (*7–10*) have recently (*11, 12*) been criticized as achieving retention of the long-range order by freezing the structure with cross-coupling agents rather than forming an equilibrium liquid crystal. The conditions for polymerization with retained liquid crystalline structure have been stated (*11*) by comparison with homotropic polymerization in crystalline structures (*12–15*). For a liquid crystalline structure the translational entropy component in the expression for the total free energy should also be included. Its magnitude is difficult to estimate; for microemulsions (*16*) the contribution of the entropic free energy is similar in magnitude to the contributions of other components of the free energy.

The fact that polymerization with retention of the liquid crystalline structure may be a rare phenomenon in lyotropic liquid crystals (*11*) encouraged us to choose an alternative route: to observe structural changes during polymerization in a lyotropic liquid crystalline matrix. We are now able to report a change from one liquid crystalline structure to another during polymerization of the amphiphilic component. To our knowledge, this is the first report of such a change. It is essential to realize that the structure obtained represents the stable conformation of the system; there is no "freezing in" of a structure—it forms spontaneously from an isotropic melt.

The components of the liquid crystal were distilled water (with 0.05*M* ammonium persulfate as an initiator) and sodium undecenoate. They formed a liquid crystal in the concentration range 47 to 59 percent sodium undecenoate (by

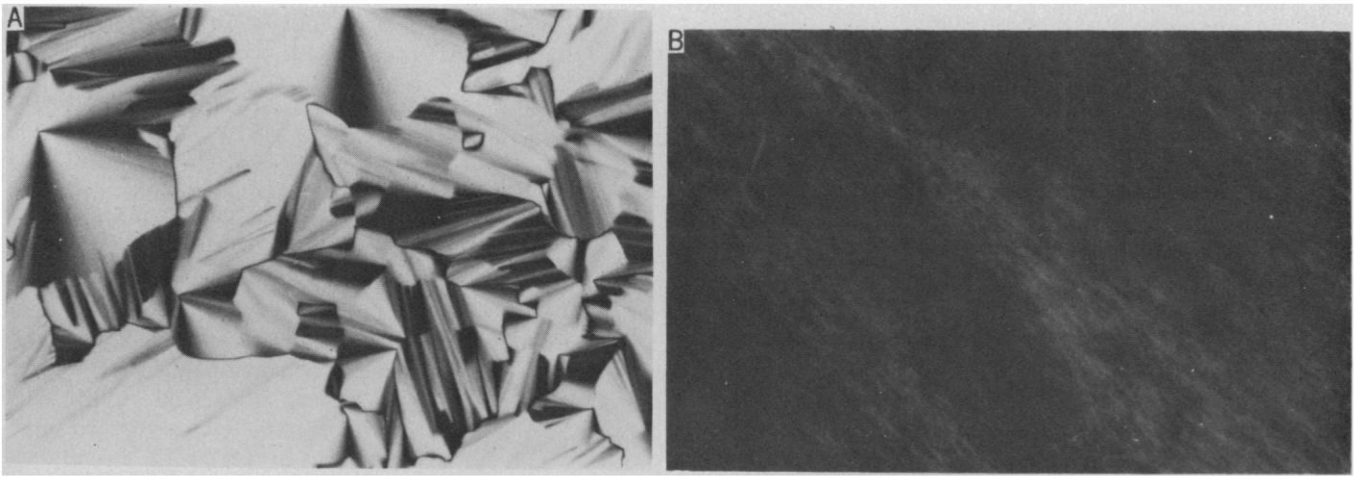

Fig. 1. (A) Optical pattern of the liquid crystalline phase before polymerization, typical of a structure of hexagonally close-packed cylinders. (B) Optical pattern of the polymerized sample.

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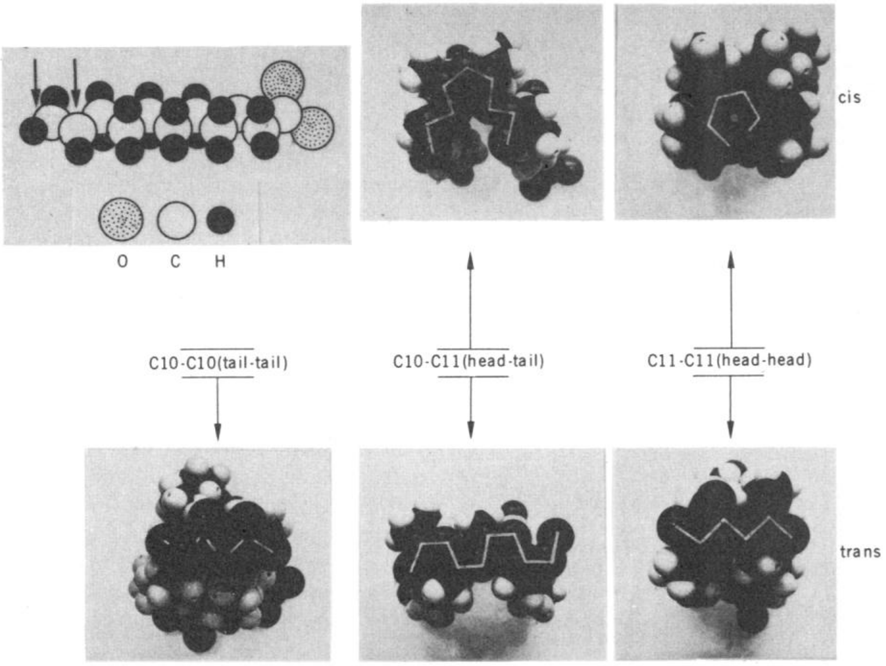

Fig. 2. Polyethylene backbone structure may accommodate head-to-head and head-to-tail conformations, with the exception of the tail-to-tail *cis* structure.

weight) at 60°C, at which temperature the polymerization took place in an N_2 atmosphere of degassed components.

The optical pattern (Fig. 1A) and low-angle x-ray reflections with radius ratios 1 : 1.71 : 2.03 indicated that the liquid crystalline structure before polymerization was a hexagonal array of close-packed cylinders (*17*). The pattern (Fig. 1A) was identical over the temperature range 20° to 75°C, leaving no doubt about the structure at 60°C before polymerization.

Structural changes during polymerization were followed at 60°C by direct microscopic observation of a sample sealed between two microscopy slides glued to spacers. Weighing of the sample during the process showed that no evaporation took place. After polymerization for 24 hours, no optical anisotropy was found; the polymerization was complete. Infrared spectra showed no double bonds in the structure. When the temperature was reduced to 20°C the optical pattern of a lamellar liquid crystalline phase appeared (Fig. 1B), and low-angle x-ray reflections of a powder from the polymerization in a sealed ampul showed distance ratios of 1 : 1/2 : 1/3 characteristic of a lamellar structure.

These results show that a lamellar structure was formed. Evidence for a lamellar liquid crystalline structure as distinguished from a crystalline structure of lamellae was the observation of one diffuse reflection at 4.5 Å characteristic of liquid hydrocarbon chains. A crystalline structure would display a series of sharp reflections in the range 3 to 4.5 Å, showing the crystalline packing of the methylene groups of the hydrocarbon chains.

The structure of the lamellar phase must be related to the fact that the molecular weight is medium; high-pressure liquid chromatography showed an average size with 270 amphiphilic units in each molecule. The backbone of the polymer is the polyethylene chain "substituted" with the remaining parts of the amphiphile chain. Molecular models showed that such a structure accommodates all head-to-tail and head-to-head configurations in the *cis* and *trans* conformations (Fig. 2) with exception of tail-to-tail in the *cis* conformation. The structure necessitates considerable bending of the hydrocarbon chains; a formal calculation of the thickness of the amphiphilic layer supports this suggestion. An expected hydrocarbon length for a normal liquid crystalline packing chain (*18*) of 14.15 Å would mean an angle of 29.4° for the hydrocarbon chain axis against the layers. There appears to be little basis for accepting such a structure. The suggested structure of a polyethylene backbone appears reasonable.

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Subicular Input from Temporal Cortex in the Rhesus Monkey

Abstract. *The subicular cortices of the primate hippocampal formation form a physical and connectional link between the cortex of the temporal lobe and the hippocampus. Their direct connections with all classes of cortex in the temporal lobe except primary sensory cortex underscore the pivotal role of these areas in the potential interplay between the hippocampal formation and the association cortices.*

Since their description nearly a century ago (*1*) the subicular cortices of the mammalian hippocampal formation (*2*) have had the vague anatomical distinction as simply the gray matter passageway interposed between the hippocampal allocortex medially and the occipitotemporal isocortex laterally. Little else has been known about these architectonically heterogeneous areas despite the fact that they reach their greatest relative size and elaboration in primates, including humans (*3*). Recently, however, anatomical results have made it increasingly clear that the subicular cortices in fact hold a pivotal position within the hippocampal formation for relaying the output of the hippo-

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