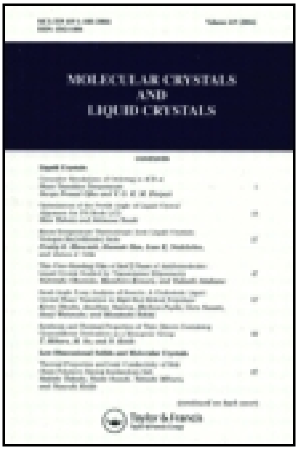

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Introduction

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Introduction

On the occasion of his 65th birthday, a one day meeting (18th June 1999) has been organized in Strasbourg in honour of Antoine Skoulios, who has contributed so much to the progress of liquid crystalline and polymer science. All the participants (P.G. de Gennes, J.C. Wittmann, D. Guillon, J. Charvolin, H. Coles, G. Hadziioannou) have recalled in their conferences the pertinent advances made by A. Skoulios during his long scientific career. At the time when he is going to retire officially, many of us thought it would be interesting to collect a series of papers in a special issue of Molecular Crystals and Liquid Crystals, in order to stress out his major contributions, and to show how these are still useful in the recent progresses of soft matter. I am quite happy that many of our colleagues have sent original papers with new results and I thank all the authors for their participation.

A. Skoulios joined the CNRS (National Scientific Research Centre) in 1955 to prepare a thesis under the supervision of V. Luzzati at the Centre for Macromolecular Science in Strasbourg. He got his PhD in 1959 on the "Structure of liquid crystalline phases obtained with pure sodium soaps and with binary mixtures of sodium soaps and hydrocarbons". He has been the leader of the X-ray group in this Centre from 1963 to 1987. Since 1987, he moved into the new Institute for Physics and Chemistry of Materials located also in Strasbourg. His main contributions are based on the concept of the amphiphatic character that he used extensively to describe the mesomorphic structures of thermotropic and lyotropic liquid crystals: soaps, polysoaps, calamitic and discotic liquid crystals. This concept has been generalized to the case of block copolymers to describe the different lamellar, columnar and cubic phases exhibited by these materials (*Mol. Cryst. Liq. Cryst.* **165** (1988) 317).

At the beginning of his carrier, he studied lyotropic liquid crystals and described the main structures of lyotropic phases of soaps by using the concepts of intramolecular segregation and independance of antagonistic molecular moieties (i.e. amphipathic character due to the difference in hydrophilic and hydrophobic interactions). He determined the local disorder of aliphatic chains and the long range order within the mesophases of these systems (*Nature* **180** (1957)

600; *Coll. Interf. Sci.* **1** (1967) 79). He also considered the thermotropic phases of amphiphiles. In this case, he found that their mesomorphic behaviour was the result of the melting, through successive steps, of the aliphatic chains and of the polar groups of the molecules. Thus, the structures of alkaline, alkaline-earth and transition metal soaps were found and described in detail : the lamellae, ribbons, cylinders or disks were found to be the fundamental structural objects (*Ann. de Physique* **3** (1978) 421). In a similar approach, the mesomorphic behaviour of calamitic liquid crystals was attributed to the amphipathic character of their molecules, due to the fact that long rigid polarizable moities and aliphatic chains prefer to segregate into different microdomains in space. The occurrence of the mesomorphic state is due to the melting of the aliphatic chains which ensures the fluidity of the mesophases, and the nature of the mesophases is determined by the degree of order of the polarizable parts (*J. de Physique* **37** (1976) 797).

During the sixties, he was involved in the pioneering work associated with the first characterizations of lyotropic and thermotropic mesophases obtained with block copolymers. He extended the concept of amphiphatic character to these systems, where the microsegregration in space of the different polymer sequences results from weak repulsive interactions (van der Waals type). The main structures are based on the stacking of lamellae (smectic) of different chemical nature, the two-dimensional organisation of cylinders (columnar) or even the three-dimensional arrangement of spheres (cubic) (in "*Advances in Liquid Crystals*", Ed. Brown, Acad. Press. **1** (1975) 169).

By comparing the structure of semi-crystalline polymers with that of virtual block copolymers containing crystalline and amorphous sequences, Dr. A. Skoulios showed it was possible to determine the folding ratio of the polymer chains within the crystalline lamellae. Moreover, he found that this ratio varies discontinuously as a function of the crystallisation temperature or of the annealing temperature. Being coiled in the melt state, the chains cannot adopt the ideal stretched conformation when the very fast process of crystallization occurs. Therefore, they crystallize only at a local scale and freeze into a folded conformation (*Koll. Zeit.* **234** (1969) 1059).

The structure of the first side-chain polymers was investigated by him when he considered polysoaps formed by polyelectrolytes carrying aliphatic side-chains. These systems exhibit lyotropic mesophases quite similar to those already described for low molecular weight soaps. Then he studied mesomorphic polymers containing mesogenic side-groups, and in particular substituted polymethacrylates. By taking into account the strongly syndiotactic nature of the polymer backbone and the steric constraints, the mesogenic side groups were found to be organised in double rows and all oriented on the same side of the backbone in the smectic phases. The macromolecules, being rather stretched, adopt the shape of very long ribbons with a width close to the side-group length and a thickness corresponding to that of a double row of mesogens (*J. de Physique* **49** (1988) 1455). For the lyotropic nematic mesophases observed with aromatic polyamides, he showed via an analysis of the kinetic variation of optical textures as a function of the polymer molecular weight, its chemical nature and its concentration in solution, that the threads in the textures correspond to a pure divergent local distortion, in agreement with the rather important rigidity factor of the polymer chains (*J. de Physique* **38** (1977) 307).

He also participated in numerous structural studies of columnar mesophases: transition metal soaps, phthalocyanine derivatives, polycatenar compounds, polydialkylsilanes and others. The structures found revealed to have the same symmetry as those already described by himself many years ago with alkaline and alkaline-earth soaps. However, it was possible now to describe in much more detail the internal structure of the columnar core: pseudo-polymeric chains of metal atoms in the case of transition metal soaps, stacking of the phthalocyanine rings normal or tilted with respect to the columnar axis, stacking of clusters of a few molecules with polycatenar compounds, gauche conformation of the silicone backbone with polydialkylsilanes (*Europhys, Lett.* **3** (1987) 67; *Liq. Cryst.* **9** (1991) 369).

More recently, A. Skoulios was very much interested in the study of liquid crystals showing hydrogen bonds and dipolar interactions. For example, in the case of N-substituted alkylammonium bromides, a tetragonal organisation of the polar heads is found within the polar sublayers when the substituents are inactive (methyl, alkyl). The tetragonal order is destroyed when the ionic heads carry only one active group, but it is reinforced when they carry two active groups, leading to the building of supramolecular assemblies ("grid" type) through hydrogen bonds (*Liq. Cryst.* **13** (1993) 201; *Liq. Cryst.* **22** (1997) 51). These last years, he has also been involved in the field of metallomesogen liquid crystals, for example of copper complexes, obtained with 2,2′-bipyridine derivatives. In this case the mesophases are no longer obtained through the classical rod-like or disk-like shaped molecules, but through supramolecular assemblies of "helicate" type, organized in columnar mesophases (*Angew. Chem. Int. Ed.* **37** (1998) 1255).

This brief overview of major achievements show how productive and remarkable the research of A. Skoulios in liquid crystals and polymers bas been during 45 years. Moreover, it should be added that A. Skoulios is the author of more than 250 publications, has supervised more than 40 PhD thesis during his career and has also been invited to many international conferences. Again, I thank all the authors for their contributions to this special issue. Finally I would like to express my gratitude to you, Antoine, for my personal training but also for the illuminating discussions we still continue to have in our laboratory.

Strasbourg, 8th June 2000
Daniel Guillon