PERIODIC DISCLINATION ARRAYS IN CARBONACEOUS MESOPHASE PITCH

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Introduction

Industrial fabrication of mesophase carbon fibers is based on the melt spinning of mesophase carbon pitch into micron-size cylindrical filaments. These fibers can attain exceptional levels of mechanical and thermal performance by appropriately controlling the final microstructure by manipulating the process conditions and the spinneret flow geometry. In various spinning experiments [3, 4, 5], it has been observed that the microstructure of mesophase filaments can be altered by such means as placing a screen mesh or perforated plate across the entry tube to the spinneret. One of the most striking observations in these experiments was the formation emergence of a regular array of $+2\pi$ and $-\pi$ disclinations at some distance below the screen under suitable flow and relaxation conditions. An example is shown in Figure 1 with a schematic map of the corresponding layer structures [4]. The layer plane structure is seen to be circumferential within each cell and aligned at the boundaries. The circumferential structure thus represents an array of $+2\pi$ disclinations, and the aligned regions at the boundaries result from the relaxation of the weld zones of the screen wire.

There has been no modelling of these complex flows to this date. Here we present some theoretical considerations leading to orderly arrays of $+2\pi$ disclinations in carbonaceous mesophase pitch flowing through a screen-mesh [2].

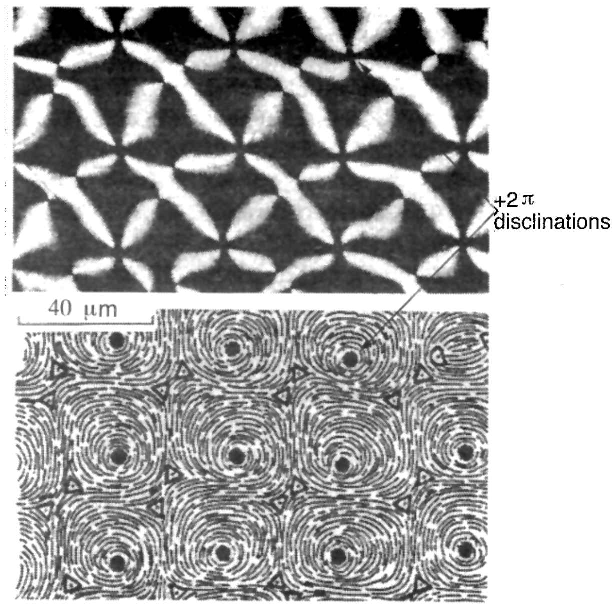

Figure 1: Regular array of disclinations formed by flow through a 200-mesh screen. Above: transverse section by crossed polars. Below: structural sketch.

Theoretical Analysis

We use Leslie-Ericksen continuum equations to describe the dynamics of mesophase pitch, modelled as a discotic liquid crystal. The flow Reynolds number based on both the mesh size and capillary diameter is near zero. Neglecting the inertia and gravity terms and with the assumption of incompressibility, the mass and momentum conservation equations reduce to

$$\nabla \cdot \mathbf{v} = 0, \quad \nabla \cdot \mathbf{T} = \nabla p$$

where p is the pressure. A convenient form of the Leslie-Ericksen viscous stress $\mathbf{T}$ and the torque balance is given [2] as

$$\mathbf{T} = \mu_1 \mathbf{n}(\mathbf{n}.\mathbf{D}.\mathbf{n})\mathbf{n} + \mu_2[\mathbf{n}(\mathbf{D}.\mathbf{n}) + (\mathbf{D}.\mathbf{n})\mathbf{n}]$$
$$+\alpha_4 \mathbf{D} + \mu_3(\mathbf{nh} - \mathbf{nn}(\mathbf{h}.\mathbf{n})) + \mu_4(\mathbf{hn} - \mathbf{nn}(\mathbf{h}.\mathbf{n}))$$
$$\text{and} \quad \gamma_1(\partial \mathbf{n}/\partial t + (\mathbf{v}.\nabla)\mathbf{n}), = \mathbf{h} \cdot (\mathbf{I} - \mathbf{nn}) + \gamma_1 \boldsymbol{\Omega} \cdot \mathbf{n}$$
$$-\gamma_2 \mathbf{D}.\mathbf{n} + \gamma_2 \mathbf{n}(\mathbf{n} \cdot \mathbf{D} \cdot \mathbf{n})$$
$$\text{where} \quad \mathbf{D} = \frac{1}{2}(\nabla \mathbf{v} + \nabla \mathbf{v}^{\mathbf{T}}), \boldsymbol{\Omega} = \frac{1}{2}(\nabla \mathbf{v} - \nabla \mathbf{v}^{\mathbf{T}})$$

and the unit vector $\mathbf{n}$ is the director giving the local direction of alignment. The coefficients are defined as $\mu_1 = (\alpha_1\gamma_1 + \gamma_2^2)/\gamma_1$, $\mu_2 = (\alpha_5\gamma_1 - \gamma_2\alpha_2)/\gamma_1$, $\mu_3 = (\alpha_2/\gamma_1)$ and $\mu_4 = (\alpha_3/\gamma_1)$ where $\alpha_1, ..., \alpha_5$ are the Leslie viscosities with $\gamma_1 = (\alpha_3 - \alpha_2)$ and $\gamma_2 = (\alpha_2 + \alpha_3)$. For discotics, $\alpha_2 > 0$ and the ratio $\alpha_3/\alpha_2 > 0$ can take values much larger than one. We assume a simple expression for the elastic molecular field, $\mathbf{h} = K\nabla^2\mathbf{n}$.

We assume the idealized screen of rectangular grid to be in the x-y plane with the grid element repeating itself to fill the entire plane of the screen so as to avoid consideration of the edge effects. We further assume that at some distance below the screen the mesophase pitch flow is strictly in the plane normal to the screen i.e. $\mathbf{v} = (0, 0, v_z(x, y))$. The spatial periodicity of the flow will be the same as that of the screen. We define a local non-dimensional Ericksen number, $Er = (\gamma d\alpha_4)/K$ where d is the mesh size and γ, a measure of the local velocity shear e.g. $\gamma = \sqrt{\mathbf{D}\cdot\mathbf{D}}$. In the region where the elastic effects are negligible ($Er >> 1$), we look for solutions with the director $\mathbf{n}$ in the x-y plane i. e. $\mathbf{n} = (n_x, n_y, 0)$. The balance equations reduce to

$$n_y\frac{\partial v_z}{\partial y} + n_x\frac{\partial v_z}{\partial x} = \mathbf{n}.\nabla\mathbf{v} = \mathbf{0} \quad (1)$$

$$\frac{\alpha_4}{2}(\frac{\partial^2 v_z}{\partial^2 x} + \frac{\partial^2 v_z}{\partial^2 y}) = \frac{dp}{dz} \quad (2)$$

In this case the flow field is independent of the director orientation and is given by the Stokes flow through a spatially periodic screen. The director orientation is also independent of the material parameters of the fluid and is given by the local shear field. The Stokes flow equations (2) are solved with the periodic boundary conditions on the velocity. We note $\partial v_z/\partial x = 0$ and $\partial v_z/\partial y = 0$ respectively on the grid centerlines parallel to y and x directions respectively.

From symmetry considerations the pitch velocity is maximum at grid center, $\partial v_z/\partial x = \partial v_z/\partial y = 0$, with vanishing shear. In the region where the shear vanishes the elastic term $\mathbf{h}$ has the dominant contribution ($Er << 1$) and the balance equations in this limit reduce to

$$\mathbf{h}\cdot(\mathbf{I} - \mathbf{nn}) = 0 \Longrightarrow \mathbf{h} = K\nabla^2\mathbf{n} = 0 \quad (3)$$

In this region of elastic dominance, the director is no longer confined to the x-y plane but escapes to the z direction corresponding to the well-known finite elastic energy solutions [1] .

$$\cot(\frac{\pi}{4} + \frac{\beta}{2}) = \frac{r}{R} \text{ and } \tan(\frac{\pi}{4} + \frac{\beta}{2}) = \frac{r}{R} \quad (4)$$

where $r = \sqrt{x^2 + y^2}$,$n_x = \cos\zeta\cos\beta$,$n_y = \cos\zeta\sin\beta$ and $n_z = \sin\beta$. More general finite elastic energy solutions to these equations corresponding to array of alternate polarity disclinations can be constructed using the theory of harmonic maps [2]. In the intermediate region, where the elastic and viscous torques balance each other the solution is determined by asymptotic matching of the two solutions.

Conclusion

The analysis reveals a class of spatially periodic solutions to Leslie-Ericksen equations. An array of $+2\pi$ disclinations, oriented along the flow direction is observed in the regions of negligible shear in the transverse plane. The requirement of transverse plane spatial periodicity of the flow results in spatially periodic variable shear leading to these disclinations. Our theoretical solutions mimick the experimental observations of disclination arrays in mesophase pitch and thus provide an understanding of the interaction of shear and elastic torques in the formation of these disclination arrays.

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