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Linear Stability Theory of Break-Up Dynamics of Nematic Liquid Crystalline Fibers

Alejandro D. Rey (*)

Department of Chemical Engineering, McGill University, Montreal, Quebec, H3A 2A7, Canada

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Abstract. — We present a new macroscopic model that describes the break-up dynamics of liquid crystalline anisotropic viscoelastic fibers. The fiber elasticity contains isotropic as well as orientation dependent surface contributions, and the anisotropic bulk viscous dissipation is described by three viscosity coefficients. For liquid crystalline fibers with molecular orientation along the fiber's axis the model predicts that capillary instabilities will break the fiber into an array of droplets, just as in the case of isotropic Newtonian fibers. The characteristic growth rate and wavelength of the instability are functions of the orientation dependent surface tension and the extensional viscosity. The liquid crystal surface elasticity tends to increase the wavelength and to decrease the growth rate of the fastest growing mode when compared to that of Newtonian fibers. Higher extensional viscosities decrease the time scale and increase the size scale of the new droplet morphology that emerges from unstable axisymmetric liquid crystalline fibers.

1. Introduction

Liquid crystalline materials have been extensively investigated in the past decades [1,2] due to their numerous industrial applications that range, according to molecular weight and mesophase type, from electrooptics [3] to fiber manufacturing [2,4]. Due to the possibility to introduce surface induce orientation, the interfacial properties of liquid crystals play a predominant role in electrooptical applications [3,5]. For example, physicochemical surface treatments exist to impart a specific molecular orientation at solid-liquid crystal interfaces [6].

More recent developments in the design of multiphase composite materials and in liquid crystalline polymer (LCP) fiber manufacturing pose new type of challenging problems involving the stability of liquid crystal fibers and the dynamics of liquid crystalline surfaces. One prominent example arises in the manufacturing of in-situ LCP polymer composites, in which a liquid crystal fiber morphology embedded in a polymer matrix arises by extruding the mixture through converging dies. Here a key question is to describe the parametric envelope that promotes the liquid crystal fiber stability, since fiber break-up leads to a droplet morphology that will not impart the desired mechanical property profile of the rod-like morphology. Another

(*) e-mail: Inaf@musicb.mcgill.ca

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area of great interest is to be able to predict the restrictions of capillary instabilities on liquid crystalline fiber manufacturing [2,4].

The frame of reference used in this work is the instability of rod-like morphologies by interfacial surface energy reduction, first considered by Lord Rayleigh, and known as the capillary instability [10]. Lord Rayleigh predicted that an infinitely long liquid cylinder will eventually break-up into spherical droplets if an infinitesimal sinusoidal perturbation of wavelength greater than the fiber's perimeter is introduced. A thermodynamic stability analysis of nematic LCP fibers was recently given [11]. The model accounted for bulk elasticity from orientation gradients and orientation surface elasticity. It was shown that for thick fibers the bulk orientation elasticity controls the fiber's stability, and the surface orientation elasticity can be ignored. For relatively thick fibers it was found that under some conditions (strong nematic elasticity and weak interfacial tension) the nematic LCP fibers may by stable. On the other hand for relatively thin fibers it is shown that the bulk orientation elasticity can be neglected and that the surface orientation elasticity controls the fiber's thermodynamic stability. For relatively thin fibers, such as those that arise during the fabrication of in-situ LCP thermoplastic composites, the theory predicts that nematic LCP fibers will be always be unstable to periodic perturbations of sufficiently long wavelength. In this paper we extend our previous analysis on liquid crystal fiber break-up by taking into account the anisotropic viscoelastic mechanisms that are involved in the dynamical evolution.

The main objective is to determine the governing parameters that control the stability of thin liquid crystalline fibers. The liquid crystal is assumed to have uniaxial nematic ordering, where the rod-like molecules are more or less parallel to each other but otherwise free to translate past each other [1]. The theory and analysis are based on the anisotropic viscoelasticity of liquid crystals. The particular objectives of this paper are:

(1) to formulate an anisotropic viscoelastic model that describes the break-up dynamics of liquid crystalline fibers;
(2) to identify the liquid crystalline elastic and viscous contributions to the morphological stability of liquid crystalline fibers;
(3) to find the characteristic time scales and selected characteristic sizes that emerge during the fiber break-up;
(4) to qualitatively compare the model predictions with experimental data [12].

The organization of this paper is as follows. In Section 2 we define the coordinate system and the state variables, derive the viscoelastic model, and present the governing equations that describe the fiber break-up dynamics. In Section 3 we present and discuss the LCP fiber stability analysis, and compare the results with the capillary instability of Newtonian fluids. Finally, parameter values arising in the theory are estimated, and the LCP fiber stability predictions are calculated and validated with existing experimental data.

2. Governing Equations

2.1. Definitions of Fiber Microstructure, Deformations, and Geometry. — In this paper we study the linear stability of a infinitely long cylindrical liquid crystal (LCP) thin axisymmetric fiber, surrounded by an inviscid fluid. The linear instability of the rod-like morphology is referred to as fiber break-up. The essence of the analysis is to describe the relevant anisotropic viscoelastic surface and bulk modes in the fiber, and to determine
Figure 1. — a) Definition of coordinate system and fiber geometry. The fiber is defined in a cylindrical coordinate system \((r, \varphi, z)\), and its axis is along \(z\). A uniform fiber has constant radius \(R_0\) and the outward surface unit normal is oriented along the radial direction. b) Schematic of uniform liquid crystalline fiber with axial texture. The thin rods within the fibers represent the average liquid crystal molecular orientation. For a uniform fiber the fiber radius is constant, the average orientation is along the fiber axis \((z\text{-direction})\), and at the surface the orientation is orthogonal to the outward surface unit normal \((N)\). c) Schematic of distorted liquid crystalline fiber with axial texture. For a distorted fiber the average orientation is along the fiber axis \((z\text{-direction})\) and at the surface the orientation is not orthogonal to the outward surface unit normal. The surface elasticity of liquid crystalline fibers is a function of the angle between the outward unit normal \((N)\) and the average orientation \(n\).

the growth characteristics of infinitesimal surface displacements. In other words, under which conditions does the fiber shown in Figure 1b evolve through that shown in Figure 1c. The list of assumptions used in the linear stability analysis will be given after we introduce the notation, and basic features of the theory.

We use Cartesian tensor notation, repeated indices are subjected to the summation convention, partial differentiation with respect to the \(j\)th spatial coordinate is denoted by a comma \((\text{i.e., } R_{ik,j} = \partial R_{ik}/\partial x_j)\). We use a cylindrical coordinate system \((r, \varphi, z)\), with the \(z\)-axis collinear with the fiber axis (see Fig. 1a).

In this paper we restrict the analysis to nematic liquid crystals. Nematics are characterized by uniaxial orientational order and positional disorder, and they are known as orientationally ordered fluids. The microstructure or orientational order is characterized by a unit vector \(n(n \cdot n = 1)\), denoted the director [1]. The director \(n\) is the average orientation of the rod-like molecules, which depending on temperature (thermotropics) or concentration (lyotropics) are more or less oriented along the director. The orientation within the liquid crystal fiber have many degrees of freedom and can range from the simple homogeneous and planar (\text{i.e., 2 dimensional}) texture to an in-homogeneous and non-planar (3 dimensional) texture. A common
fiber texture that is widely observed in many of the applications that motivate this research is the axial texture, where all the directors point along the fiber axis. Axial fiber textures are fairly common because they represent the lowest elastic energy state, since the director is spatially homogeneous. In addition, whenever these fibers arise during stretching deformations of droplets, the directors tend to align along the stretching direction, which coincides with the fiber axis. Other fiber textures, such as onion or radial textures [13] are beyond the scope of this paper. A side and front view schematic of the undeformed axisymmetric liquid crystalline fiber with an axial texture is shown in Figure 1b.

To identify a nematic polymer fiber we need to define, as for isotropic fibers, its shape. Here we restrict the analysis to infinitely long fibers with circular cross sections of radius \( \rho \). [All the solutions arising in this paper have azimuthal symmetry.] Figure 1c shows a liquid crystalline fiber with surface distortions. Thus at any time "t" the shape is determined by the possible variation of the radius \( \rho \) with axial distance \( z \):

\[
\rho(z, t) = a + \xi(z, t).
\]

The small displacement \( \xi \) is expressed in terms of Fourier modes:

\[
\xi(z, t) = \sum_k \xi_k e^{ikz+\alpha t}
\]

where \( \alpha \) is the growth rate and \( \xi_k \) is the amplitude of the \( k \)th mode. For small displacements the principal curvatures of the fiber at time "t" and the unit outward normal \( N \) are given by [14]:

\[
R_r\theta = \sqrt{1 + \xi'^2} R \approx R = a + \xi
\]  

(3a)

\[
R_zr = \left( \frac{1 + \xi'^2}{-\xi''} \right)^{3/2} \approx -\frac{1}{\xi''}
\]  

(3b)

\[
N_r = \left( 1 + \xi'^2 \right)^{1/2}; \quad N_\theta = 0; \quad N_z = -\xi \left( 1 + \xi'^2 \right)^{-1/2}
\]  

(3c)

The assumptions and restrictions imposed on the linear analysis made in this paper are as follows:

1) the single liquid crystalline fiber is embedded in an inviscid fluid;

2) incompressible and isothermal conditions hold;

3) the temperature or concentration corresponds to the nematic phase of a nematic liquid crystal;

4) the shapes of the fibers are known a priori, and have azimuthal symmetry;

5) the director field is constant and given by \( n = (0, 0, 1) \), regardless of fiber shape. This assumption follows from the thin fiber restriction stated in the objective enumerated on Section 1. The total free energy \( F_t \) of nematic liquid crystalline cylindrical fiber is given by:

\[
F_t = 2\pi KL \int F_r r dr dz + 2\pi RGL \int F_\theta dz
\]  

(4)
6) the analysis is restricted to the linear regime. The linear stability analysis predicts that when a fiber is unstable it transforms into a train of equally sized droplets. In practice it is observed in isotropic viscous and viscoelastic jets that smaller satellites drops appear. To capture the emergence of satellites drops as well the intermediate and late stage (pinching stage) of the fiber instability, a nonlinear analysis of the full set of linear momentum and director balance equations that involves shape and director distortions must be performed.

Situations involving microstructure or geometry not restricted as above lie beyond the range of validity of the present analysis, and beyond the scope of this paper.

2.2. GOVERNING EQUATIONS. — The linear momentum balance equation in cylindrical \((r, \theta, z)\) coordinates are to first order given by [15]:

\[
\begin{align*}
\rho \frac{\partial v_r}{\partial t} &= - \frac{\partial p}{\partial r} + \frac{1}{r} \frac{\partial (rt_{rr})}{\partial r} + \frac{\partial (t_{rz})}{\partial z} - \frac{t_{\theta \theta}}{r} \\
\rho \frac{\partial v_z}{\partial t} &= - \frac{\partial p}{\partial z} + \frac{1}{r} \frac{\partial (rt_{rz})}{\partial r} + \frac{\partial t_{zz}}{\partial z}
\end{align*}
\]  

(5a) (5b)

where \(t\) is the extra stress tensor, \(p\) is the pressure, \(v\) is the velocity vector, and \(\rho\) is the density. The equation of continuity is given by [15]:

\[
\frac{\partial v_z}{\partial z} + \frac{1}{r} \frac{\partial (rv_r)}{\partial r} = 0.
\]

(6)

The constitutive equation for the extra stress tensor \(t\) used in this work is that of the Transverse Isotropic Fluid (TIF) theory of Ericksen [16]. In this viscous model the extra stress tensor \(t\) is symmetric and given by:

\[
t^v = \eta_3 (A : nn)nn + 2\eta_2 A + 2(\eta_1 - \eta_2)(A \cdot nn + nn \cdot A)
\]

(7)

where \(A\) is the symmetric traceless rate of deformation tensor given by

\[
A_{ij} = A_{ij} = \frac{1}{2} \left( \frac{\partial v_i}{\partial x_j} + \frac{\partial v_j}{\partial x_i} \right).
\]

(8)

Here we are using the nomenclature of [17]. When the viscosity coefficient \(\eta_1\) is equal to \(\eta_2\) and when \(\eta_3 = 0\) the theory reduces to the isotropic Newtonian fluid theory. The dynamics of the director is given by:

\[
\frac{\partial n}{\partial t} + (v \cdot \nabla)n - W \cdot n = \lambda(A \cdot n - A : nn)
\]

(9)
where $\lambda$ is a dimensionless parameter and $W$ is the vorticity tensor:

$$W_{ij} = -W_{ji} = \frac{1}{2} \left( \frac{\partial v_i}{\partial x_j} - \frac{\partial v_j}{\partial x_i} \right).$$  \hspace{1cm} (10)

The kinematic boundary condition required to integrate the momentum balance equations is given by:

$$v_r = \frac{\partial \xi}{\partial t} + \frac{\partial \xi}{\partial z} v_z, \quad r = a + \xi$$  \hspace{1cm} (11)

and indicates that fluid does not cross the free surface. The first dynamic boundary condition corresponds to vanishing shear stress at the free surface [14]:

$$(T \cdot N) \cdot k = 0$$  \hspace{1cm} (12)

where $T$ is the total stress tensor: $T = -p \delta + t$. The second dynamic boundary conditions is the Young-Laplace equation for a liquid crystal free surface, obtained using a balance of normal forces, and reads [18]:

$$-(T \cdot N) \cdot \nabla \frac{\delta V}{\delta \xi} = \frac{\delta F_s}{\delta \xi}$$  \hspace{1cm} (13)

where $V$ is the total volume of the fiber and $F_s$ is the total surface energy. The surface energy density $f_s$ used in this paper is given by the classical and well-known Rapini-Papoular equation (1):

$$f_s = \tau_1' - \frac{\tau_2}{2} (\nu \cdot n)^2$$  \hspace{1cm} (14)

where $\nu$ is the preferred surface orientation, $\tau_1'$ is the isotropic contribution to the surface tension, and $\tau_2$ is the orientation dependent contribution. When $n$ is parallel to $\nu$ the surface energy density is a minimum, and when it is orthogonal it is a maximum. For axisymmetric fibers and with $\nu$ along the fiber axis, the first order terms in the surface energy density are:

$$f_s = \tau_1 + \frac{\tau_2}{2} (\frac{\partial \xi}{\partial z})^2$$  \hspace{1cm} (15)

where $\tau_1 = \tau_1' - \tau_2/2$. When $\tau_2$ is equal to zero the surface tension is independent of orientation, as in an isotropic Newtonian fluid. This reference state will be used to contrast the stability of the liquid crystalline fiber with the classical Newtonian fluid fiber. In partial summary, the theory used here captures the underlying orientation of the liquid crystal fiber, and describes the fiber as an anisotropic viscoelastic medium, with viscous anisotropy in the bulk as well as surface elastic anisotropy.

3. Dynamics of Liquid Crystal Fiber Break-up

In this section we derive and solve the equations that describe the break-up dynamics of liquid crystalline fibers. The break-up dynamics arises from the appearance of symmetric surface waves. A classical [18] simplifying assumption adopted here is that the wavelength of the surface waves are much longer than the fiber radius.

In the long-wavelength approximation the continuity equation shows that $v_z \gg v_r$, and therefore the axial velocity $v_z$ is the only significant velocity component. In addition since the liquid crystal fiber has a free surface, the radial dependence of $v_z$ can be ignored. Thus the velocity vector simplifies to: $v(z, t) = (0, 0, v_z(z, t))$. 


Using the orientation field \( \mathbf{n} = (0, 0, 1) \) corresponding to average molecular alignment along the fiber axis, the linear momentum balance equations and the TIF stress constitutive equation give:

\[
\begin{align*}
\rho \frac{\partial v_z}{\partial t} &= \frac{\partial}{\partial z} \left( T_{zz} - \eta_2 \frac{\partial v_z}{\partial z} \right) \quad (16a)
\frac{\partial p}{\partial r} &= 0. \quad (16b)
\end{align*}
\]

To make further progress the stress boundary condition (Eq. (13)) along the surface normal (Young-Laplace equation) needs to be specified. As shown in the Appendix the Young-Laplace equation is given by:

\[
[(p\delta - t) \cdot \mathbf{N}] \cdot \mathbf{N} = \tau_1 \left( \frac{1}{R_{rz}} + \frac{1}{R_{r\theta}} \right) + \tau_2 \left( \frac{1}{R_{r\theta}} \right) \quad (17)
\]

which properly reduces to the Newtonian fluid case when \( \tau_2 = 0 \), which in terms of the displacement reduces to:

\[
p \equiv p_r = \frac{\tau_1}{a} - \frac{\tau_1}{a^2} \left( \xi + (1 + \tau) \frac{\partial^2 \xi}{\partial z^2} \right) \quad (18)
\]

where \( \tau = \tau_2/\tau_1 \). A useful expression for the pressure \( p \) appearing in the above equation is given the average of the normal stresses:

\[
p = -\frac{1}{3} (T_{zz} + T_{rr} + T_{\theta\theta}) = -\frac{1}{3} (T_{zz} + 2p_r). \quad (19)
\]

Replacing the above expression for the pressure in \( T_{zz} = -p + t_{zz} \), and using the expression of \( t_{zz} \) given by the TIF model we find that \( T_{zz} \) is given by:

\[
T_{zz} = p_r + (2\eta + \eta_2) \frac{dv_z}{dz} \quad (20)
\]

where \( \eta = 3\eta_3/4 + 3\eta_1 - 2\eta_2 \). For later reference it is useful to note that the extensional viscosity \( \eta_e \) is given by [16]:

\[
\eta_e = 3\eta_2 + 4(\eta_1 - \eta_2) + \eta_3 \quad (21)
\]

and when \( \eta_1 = \eta_2 \) and \( \eta_3 = 0 \) it properly reduces to the Troutonian viscosity: \( \eta_e = 3\eta_2 \). Expressing \( \eta \) in terms of \( \eta_e \) we find:

\[
\eta = 3\eta_e/4 - 5\eta_2/4 \quad (22)
\]

which identifies the central role of the extensional viscosity in the stability of liquid crystalline fibers with axial microstructure (i.e., \( n_z = 1 \)). The viscosity \( \eta \) thus represents the excess of the extensional viscosity over the Newtonian viscosity, and for Newtonian fluids this excess is just \( \eta_2 \).

Finally, replacing the expression for \( T_{zz} \) into equation (16a) we find:

\[
\rho \frac{\partial v_z}{\partial t} = \frac{\partial p_r}{\partial z} + 2\eta \frac{\partial^2 v_z}{\partial z^2}. \quad (23)
\]

A relation between displacement \( \xi \) and axial velocity \( v_z \) is obtained using the equation of continuity (18):

\[
a \partial v_z/\partial z = -2\partial \xi/\partial t. \quad (24)
\]
Using the above equation, together with the expression for $p_r$ (see Eq. (18)) in equation (23) finally gives the governing differential equation for the displacement:

$$\frac{\partial^2 \xi}{\partial t^2} - 2\frac{\eta}{\rho} \frac{\partial^3 \xi}{\partial z^2 \partial t} + \frac{\tau_1}{a^2} \frac{\partial^2}{\partial z^2} \left[ \xi + a^2(1 + \tau) \frac{\partial^2 \xi}{\partial z^2} \right] = 0$$

(25)

which properly reduces to the Newtonian case if viscoelastic anisotropy is neglected: $\tau = 0$, $\eta_1 = \eta_2$, and $\eta_3 = 0$. Replacing a linear combination of spatially harmonic modes:

$$\xi(z, t) = \sum_k \xi_k e^{ikz + \alpha t}$$

(26)

gives the following expression for the growth rate $\alpha$:

$$\alpha = -\frac{\eta}{\rho} k^2 + \sqrt{\left(\frac{\eta}{\rho} k^2\right)^2 + \frac{\tau_1 k^2}{2\rho a} (1 + (1 + \tau)(ka)^2)}$$

(27)

which properly reduces to the Newtonian case if viscoelastic anisotropy is neglected [14]. The growth rate of the fastest growing mode $\alpha_{\text{max}}$ is:

$$\alpha_{\text{max}} = \frac{1}{\sqrt{\frac{8a^3(1+\tau)\rho}{\tau_1} + \frac{4a\eta}{\tau_1}}}$$

(28)

The characteristic size of the liquid crystal droplets that are expected to form from the fiber break-up is found to be:

$$\lambda = \frac{2\pi}{k_{\text{max}}} = 2\pi \sqrt{\frac{2(1 + \tau)a^2 + \sqrt{\frac{8a^3(1 + \tau)\eta^2}{\rho\tau_1}}}{}}$$

(29)

where $k_{\text{max}}$ is the wave vector corresponding to the maximum growth rate $\alpha_{\text{max}}$.

The specific contributions arising from liquid crystallinity arise from the surface anisotropic elastic term $\tau$, and from the excess viscosity coefficient $\eta$; referring back to equations (28, 22) the role of the extensional viscosity on $\alpha_{\text{max}}$ and $\lambda$ is made clear. A number of conclusions can be drawn from the expressions for the maximal growth rate and droplet size. For highly viscous materials, such as polymeric liquid crystals, the maximal growth rate is unaffected by the orientation dependent surface tension and decreases with increasing extensional viscosity (see Eq. (22)):

$$\alpha_{\text{max}} = \frac{\tau_1}{4a\eta}.$$  

(30)

The characteristic droplet size increases with increasing surface orientation elasticity and with the extensional viscosity:

$$\lambda = \frac{2\pi}{k_{\text{max}}} = 2\pi \left[ \frac{8a^3(1 + \tau)\eta^2}{\rho\tau_1} \right]^{1/4}$$

(31)

On the other hand neglecting viscous effects, the maximal growth rate is a decreasing function of the orientation surface elasticity:

$$\alpha_{\text{max}} = \sqrt{\frac{\tau_1}{8a^3(1 + \tau)\rho}}$$

(32)
while the droplet size is an increasing function of the surface elasticity:

\[ \lambda = \frac{2\pi}{k_{\text{max}}} = 2\pi a \sqrt{2(1 + \tau)}. \]  

Finally it is of interest to give estimates using reported physical property data. For a typical low molar mass nematic liquid crystals para-azoxanisol (PAA) at 135 °C, we find that \( \tau_1 = 38 \text{ dynes/cm}, \rho = 1.168 \text{ g/cc}, \eta = 14.84 \text{ cp} \) [19]. Assuming that \( \tau = 1.1 \), for a fiber of radius \( a = 1 \mu m \), the droplet size is \( \lambda \approx 28 \mu m \), and for a fiber of radius \( a = 10 \mu m \), the droplet size is \( \lambda \approx 133 \mu m \). On the other hand, for a typical lyotropic polymeric liquid crystal such as poly-benzylglutamate (PBG) we find \( \eta = 21.67 \text{ poise} \) [20], and assuming that \( \tau_1 = 1 \text{ dyne/cm} \) [21], \( \rho = 1 \text{ g/cc} \), and \( \tau = 1.1 \), we find that if \( a = 1 \mu m \), the droplet size is \( \lambda \approx 75 \text{ mm} \), and if \( a = 10 \mu m \) the droplet size is \( \lambda \approx 276 \text{ mm} \). Although the analysis presented here is for a liquid crystalline fiber surrounded by a inciscid fluid these results are in qualitative agreement with experimental data of [12] (see Figs. 2, 4, and 5 of Verhoogt et al. (1993)), obtained using Vectra 900, a thermotropic liquid crystalline polymer, blended with Kraton G1650, which is a thermoplastic elastomer. The experiments show that annealing of isolated LCP fibers as well as fibers embedded in the thermoplastic elastomer matrix results in fiber break-up behavior that resembles that of Newtonian fibers [12].

The qualitative agreement with the one available experimental observation described here is not sufficient to establish the general validity of the theory presented here. To further test the general validity of the theory presented here additional experimental data are necessary for other nematic materials in a range of fiber sizes and for a range of different temperatures, as well as a way to measure the sensitivity of nematic parameter \( \tau \) (defined below Eq. (18)) to temperature, to the molecular weight of the nematic, among other parameters. It should also be noted that no elastic forces have been included in the linear momentum balance equation, but for polymeric fibers under certain circumstances the response may be viscoelastic.

4. Conclusions

A model that describes the break-up of a liquid crystalline anisotropic viscoelastic fiber has been presented. The fiber elasticity contains isotropic as well as orientation dependent surface contributions, and the viscous dissipation is described by three viscosity coefficients. For liquid crystalline fibers with axial molecular orientation the model predicts that capillary instabilities will break the fiber into an array of droplets, just as in the case of Newtonian fibers. For liquid crystalline fibers with axial orientation the characteristic growth rates of the instability and droplet sizes are functions of the orientation dependent surface tension and the extensional viscosities. The elasticity due to liquid crystallinity tends to increase the wavelength and decrease the growth rate of the fastest growing mode when compared to Newtonian fibers. The extensional viscosity is a basic rheological property that also affects the time and size scales of the new morphology that emerges from unstable axisymmetric liquid crystalline fibers.

Acknowledgments

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Appendix

In this appendix we derive equation (17). The starting point is a balance of forces in the normal direction to the free surface:

\[-( \mathbf{T} \cdot \mathbf{N}) \cdot \mathbf{N} \frac{\delta V}{\delta \xi} = \frac{\delta F_s}{\delta \xi} \]  \hspace{1cm} (13)

In the presence of a small surface displacement $\xi$ on a fiber of radius $a$, the variation of the volume $\delta V$ is (22):

\[\delta V = \left[ \frac{1}{(a + \xi)} \right]^{-1} \delta A. \]  \hspace{1cm} (A.1a)

Using the Rappini-Papoular surface energy density equation (15) we find:

\[f_s = \tau_1 + \frac{\tau_2}{2} (\mathbf{N} \cdot \mathbf{n})^2 = \tau_1 + \frac{\tau_2}{2} \cos^2 \psi = \tau_1 + \frac{\tau_2}{2} \left( \frac{\partial \xi}{\partial z} \right)^2 \]  \hspace{1cm} (A.2)

and the total surface free energy $F_s$ is:

\[F_s = 2\pi \int \left[ \tau_1 + \frac{\tau_2}{2} (\mathbf{N} \cdot \mathbf{n})^2 \right] \left[ a + \xi + \frac{a}{2} \left( \frac{\partial \xi}{\partial z} \right)^2 \right] \, dz. \]  \hspace{1cm} (A.3)

Taking the first variation of the total surface free energy $F_s$ gives:

\[\delta F_s = 2\pi \int \left[ \tau_1 + \frac{\tau_2}{2} (\mathbf{N} \cdot \mathbf{n})^2 \right] [1 - (a + \xi)\xi'] \delta \xi \, dz + 2\pi \int \tau_2 [- (a + \xi)\xi''] \delta \xi \, dz. \]  \hspace{1cm} (A.4)

Replacing the variational derivatives into the force balance equation (13), retaining the leading terms, and making use of the expressions for the principal radius of curvatures (3a, 3b) we get equation (17).

References