Generalized cholesteric permeation flows

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(Received 18 June 2001; published 15 January 2002)

The permeation flow equations of cholesteric liquid crystals are derived using a decoupled formulation of the Leslie-Ericksen equations. The formulation sheds light on the role of Ericksen elastic stresses in permeation flows. The Darcy flow regime is shown to emerge in the absence of velocity gradients. The permeation flow equations are generalized to gravity driven flow and used to analyze a free-boundary film flow over an inclined plane.

DOI: 10.1103/PhysRevE.65.022701 PACS number(s): 64.70.Md, 47.20.Ft, 47.20.Hw, 47.50.+d

I. CHOLESTERIC ORDERING, PERMEATION FLOWS, AND LESLIE-ERICKSEN EQUATIONS

Cholesteric liquid crystals (CLCs) are biological and synthetic materials whose average macroscopic molecular orientation $\mathbf{n}^o$ exhibits a twist distortion in the direction normal to the molecules [1] known as the cholesteric helix orientation $\mathbf{p}$. Examples of CLCs are solutions of DNA [2], polypeptides, and hydroxypropylcellulose [1]. Previous work on the shear flow of CLCs has considered $\mathbf{p}\|\mathbf{v}$ [1,3], along $\nabla\mathbf{v}$ [4,5], and along the curl $\mathbf{v}$ [6–9], where $\mathbf{v}$ is the velocity. Recent reviews can be found in the literature [1,10–12]. Studies of CLCs under shear have shown that the materials are highly dependent on the angle between $\mathbf{p}$ and $\mathbf{v}$. Capillary Poiseuille flow when the helix is along the velocity direction leads, at small pressure drops, to a highly viscous permeation flow. The main characteristic of the permeation mode is its high viscosity [13]. While for all other flow geometries CLCs have apparent viscosities of the same order of magnitude as nematics, for permeation flow, their ratio is of the order 10^6. The high viscosity of permeation pressure-driven flow in capillaries has been explained using dissipation arguments as well as the Leslie-Ericksen (LE) equations for CLCs [1,3]. In these works, the analysis is based on the couplings between the linear momentum and director torque balance equations, but a systematic discussion of the role of elastic stresses has not been presented. Here, we present a different and efficient decoupled formulation that directly leads to the kinematics of permeation flow. The formulation is based on expressing the gradient of the elastic stresses that appear in the linear momentum equation in terms of viscous torques that appear in the director torque balance equation. The driving forces are generalized to include pressure-driven flow as well as gravity-driven flow. Last, the formulation is applied to a free-boundary film flow, where the system size is unknown and where the parametric conditions lead to a crossover from Newtonian film flow to non-Newtonian permeation film flow.

The equilibrium state of director orientation $\mathbf{n}^o(z)$ that represents the average molecular orientation of CLCs is

$$\mathbf{n}^o(z)=(\cos\theta_o(z),\ \ \sin\theta_o(z),\ \ \ 0), \tag{1a}$$

$$\theta_o=\frac{2\pi z}{P_o}=q_o z, \tag{1b}$$

where "o" denotes equilibrium, θ_o is the orientation angle at equilibrium, $P_o(q_o)$ is the equilibrium pitch (wave vector) of the helix, or distance in which the director rotates by 2π radians; we only consider a right-handed helix ($q_o>0$). In contrast to nematics, CLCs have an intrinsic length scale given by the equilibrium pitch P_o. This means that at length scales shorter than P_o, a CLC is similar to a nematic, but at length scales longer than P_o, a cholesteric is a layered liquid similar to a smectic-A liquid crystal. The Leslie-Ericksen (LE) balance equations for cholesteric liquid crystals, using cartesian tensor notation, are [1]

$$v_{i,i}=0, \tag{2a}$$

$$\varrho\dot{v}_i=\varrho g_i+t^{\mathrm{ve}}_{ji,j}-p_{,j}+\delta_{ji}, \tag{2b}$$

$$\sigma n_i=h^{\mathrm{me}}_1+h^{v}_i\ . \tag{2c}$$

The fluid is assumed to be incompressible; ρ is the density and p is the pressure. The superposed dot denotes the material time derivative. The inertia of the director is neglected. The mechanical quantities appearing in the LE theory are defined as follows: ρg_i: gravitational force per unit volume, t^{ve}_{ij}: viscoelastic stress tensor, h^{me}_i: magnetoelastic molecular field, h^{v}_i: viscous molecular field, σ is a Lagrange multiplier due to the director unit length restriction: $\mathbf{n}\cdot\mathbf{n}=1$. In the absence of temperature and the constitutive, equations are given by

$$\hat{\mathbf{t}}=\mathbf{t}^{\mathrm{ve}}+\mathbf{t}^{E}, \tag{3a}$$

$$\mathbf{t}^{\mathrm{ve}}=\alpha_1\mathbf{nn}:\mathbf{Ann}+\alpha_2\mathbf{nN}+\alpha_3\mathbf{N_n}+\alpha_4\mathbf{A}+\alpha_5\mathbf{nn}\cdot\mathbf{A}+\alpha_6\mathbf{A}\cdot\mathbf{nn}, \tag{3b}$$

$$\mathbf{t}^{E}=-\frac{\partial f_d}{\partial\nabla\mathbf{n}}\cdot(\nabla\mathbf{n})^T, \tag{3c}$$

$$\mathbf{h}^{v}=\gamma_1\mathbf{N}+\gamma_2\mathbf{A}\cdot\mathbf{n}, \tag{3d}$$

$$\mathbf{h}^{\mathrm{me}}=-\frac{\delta F}{\delta\mathbf{n}}, \tag{3e}$$

$$2\mathbf{A}=\nabla\mathbf{v}+(\nabla\mathbf{v})^T, \tag{3f}$$

$$\mathbf{N}=\frac{\partial\mathbf{n}}{\partial t}+\mathbf{v}\cdot\nabla\mathbf{n}+\mathbf{W}\cdot\mathbf{n}, \tag{3g}$$

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$$\mathbf{W}=[\boldsymbol{\nabla}\mathbf{v}-(\nabla\mathbf{v})^T]/2, \tag{3h}$$

$$F=\int (f_d+f_m)dV, \tag{3i}$$

$$2\,f_d=K_{11}(\boldsymbol{\nabla}\cdot\mathbf{n})^2+K_{22}(\mathbf{n}\cdot\boldsymbol{\nabla}\times\mathbf{n}+q_o)^2+K_{33}(\mathbf{n}\times\boldsymbol{\nabla}\times\mathbf{n})^2, \tag{3j}$$

$$f_m=-\frac{\chi_a}{2}(\mathbf{n}\cdot\mathbf{H})^2, \tag{3k}$$

$$\gamma_1=\alpha_3-\alpha_2 \tag{3l}$$

$$\gamma_2=\alpha_3+\alpha_2, \tag{3m}$$

where $\mathbf{t}^{\mathrm{ve}}$ is the viscoelastic extra stress tensor, $\mathbf{t}^E$ is the elastic Ericksen stress, the viscosities $\{\alpha_i, i=1,...,6\}$ are the Leslie coefficients, $\delta F/\delta\mathbf{n}$ denotes the functional derivative of the free energy F, $\mathbf{A}$ is the rate of deformation tensor, $\mathbf{N}$ is the director Jaumann derivative, $\mathbf{W}$ is the vorticity tensor, f_d is the Frank free-energy density, $\{\mathbf{K}_{ii}, ii=11,22,33\}$ are the Frank elasticity coefficients for splay, twist, and bend, f_m is the magnetic energy density, χ_a is the anisotropic magnetic susceptibility, γ_2 is the irrotational torque coefficient, and γ_1 is the rotational viscosity. In the Leslie theory of CLCs, the pitch-dependent internal time τ_i and length scales ℓ_i are

$$\tau_i=\frac{\gamma_1 q_o^{-2}}{K}, \tag{4a}$$

$$\ell_i=\frac{1}{q_o}, \tag{4b}$$

while the external time and length scales are

$$\tau_e=\tau=\frac{\gamma_1 H^2}{K}, \tag{5a}$$

$$\ell_e=H, \tag{5b}$$

where H is the characteristic system size. The ratio of orientation time scale to an imposed flow time scale is known as the Ericksen number. Since CLCs have two time scales, the two dimensionless Ericksen numbers that characterize the ratio of orientation to flow time scales are

$$E_i=\frac{\gamma_1 q_o^{-2}\dot{\gamma}}{K}, \tag{6a}$$

$$E=\frac{Y_1 H^2\dot{\gamma}}{K}, \tag{6b}$$

where $\dot{\gamma}$ is the characteristic deformation rate, and $K=(K_{11}+K_{22}+K_{33})/3$ is the average Frank elastic constant. The internal Ericksen number E_i gives the ratio of the internal time scale to the flow time scale, while the external Ericksen number E gives the ratio of the external time scale to the flow time scale. In actual experiments [1] it is found

$$\tau_i \ll \tau_e, \tag{7}$$

which means that an imposed weak flow will only affect the global orientation but not the pitch. In this report, we define weak flow when E_i is of order of unity. When $\ell_i \ll \ell_e$, the presence of the large parameter $(\Lambda=q_o H \gg 1)$ in the LE equations signals the existence of boundary layer behavior.

Permeation CLC flow in a narrow capillary is obtained when the wall orientation is compatible with the helix structure and when the imposed pressure drop is low. The ideal permeation flow mode of a CLC emerges when the helix is along the flow (z) direction and the following kinematic and orientation conditions are satisfied [1]

$$\mathbf{W}=\mathbf{A}=\mathbf{0}, \tag{8a}$$

$$\mathbf{n}=\mathbf{n}_o, \tag{8b}$$

$$\frac{\partial\mathbf{n}}{\partial t}=0, \tag{8c}$$

$$\mathbf{N}=(\mathbf{v}\cdot\boldsymbol{\nabla})\mathbf{n}_o=v_z\frac{d\mathbf{n}_o}{dz}. \tag{8d}$$

Thus in the ideal permeation flow, the velocity gradient is zero, the helix is undistorted, and the director Jaumann derivative is equal to the convection of orientation. The convected orientation rotates with an angular velocity that exactly matches the pitch of the helix, dissipating energy due to the director rotation, and giving rise to a large apparent viscosity. The ideal permeation flow neglects the boundary layer of thickness q_o^{-1} in which viscous effects are not negligibly. Under these restrictions, the apparent viscosity η_{app} for an ideal steady Poiseuille permeation CLC capillary flow under pressure gradient Δp, in a capillary of radius R is [1]

$$\eta_{\mathrm{app}}=\tfrac{1}{8}\gamma_1(q_o R)^2 . \tag{9}$$

Using [1], the following actual values: $R=300\,\mu$m and $q_o=10^{-5}\,\mathrm{cm}^{-1}$, the apparent viscosity has a magnitude of a nematic rotational viscosity times a factor of 10^6, as measured by Sakamoto, Porter, and Johnson [13].

II. FORMULATION OF THE LESLIE-ERICKSEN LINEAR MOMENTUM BALANCE EQUATION FOR CHOLESTERIC PERMEATION FLOW

Next, we give a detailed analysis of generalized permeation flow, taking into account boundary layers, using a decoupled formulation. The decoupled formulation aims at finding permeation flow kinematics without resorting to the use of the director torque balance equation. In addition, the formulation sheds light on the role of the Ericksen elastic stresses. The spatial gradient of the director-dependent energy density, $f=f_m+f_d$, is given by

$$\nabla f=\frac{\partial f}{\partial\mathbf{n}}\cdot(\boldsymbol{\nabla}\mathbf{n})^T+\frac{\partial f}{\partial\boldsymbol{\nabla}\mathbf{n}}:(\nabla\boldsymbol{\nabla}\mathbf{n})^T. \tag{10}$$

Using this expression in Eq. (3c), the divergence of the Ericksen stress tensor is shown to be

$$\boldsymbol{\nabla}\cdot\mathbf{t}^E=-(\boldsymbol{\nabla}f)+\frac{\partial f}{\partial\mathbf{n}}\cdot(\nabla\mathbf{n})^T-\left(\nabla\cdot\frac{\partial f}{\boldsymbol{\nabla}\mathbf{n}}\right)\cdot(\boldsymbol{\nabla}\mathbf{n})^T. \tag{11}$$

Taking the product of the director torque balance Eq. (2c) with the director gradient tensor $(\boldsymbol{\nabla}\mathbf{n})^T$ we find

$$\mathbf{h}^{\mathrm{me}}\cdot(\nabla\mathbf{n})^T+\mathbf{h}^v\cdot(\nabla\mathbf{n})^T=-\frac{\partial f}{\partial\mathbf{n}}\cdot(\nabla n)^T+\left(\nabla\cdot\frac{\partial f_d}{\partial\nabla\mathbf{n}}\right)\cdot(\nabla\mathbf{n})^T, \quad (12)$$

and hence the following general relation between the Ericksen elastic forces, energy gradients, and viscous molecular field is established:

$$\nabla\cdot\mathbf{t}^E=-(\nabla f)-\mathbf{h}^v\cdot(\nabla\mathbf{n})^T=-(\nabla f)-\left(\gamma_1\frac{\partial\mathbf{n}}{\partial t}+\mathbf{v}\cdot\nabla\mathbf{n}+\mathbf{W}\cdot\mathbf{n}+\gamma_2\mathbf{A}\cdot\mathbf{n}\right)\cdot(\nabla\mathbf{n})^T. \quad (13)$$

Equation (13) is valid for any velocity and director fields. Equation (13) shows the conditions under which the Ericksen elastic forces may play an important role: (i) when the gradients of the Frank energy are not zero, $|(\nabla f)|\neq 0$, and/or (ii) when the $|\mathbf{h}^v\cdot(\nabla\mathbf{n})^T|\neq 0$. An example, of the former is one-dimensional (1D) permeation flow of a CLC with wave-vector q_o along the z direction and velocity field $\mathbf{v}=(0,0,v_z)$, the leading-order term in the Ericksen force in the flow direction is

$$(\nabla\cdot\mathbf{t}^E)\cdot\mathbf{k}=-\mathbf{h}^v\cdot(\nabla\mathbf{n})^T\cdot\mathbf{k}=-\gamma_1(\mathbf{v}\cdot\nabla\mathbf{n})\cdot(\nabla\mathbf{n})^T=-\gamma_1 q_o^2 v_z, \quad (14)$$

showing that the elastic Ericksen force is due to the convection of orientation $(\mathbf{v}\cdot\nabla\mathbf{n})$. Next, we consider the linear momentum balance equation and replace the gradient of the Ericksen stress with the right hand side of Eq. (13). Neglecting inertia, the linear momentum balance Eq. (2b) then becomes

$$\mathbf{v}(\mathbf{x})=\frac{\Phi}{\gamma_1}:[-\nabla(p+f_d+f_m+f_g)+\nabla\cdot\mathbf{t}^{\mathrm{ve}}-(\mathbf{h}^v-\gamma_1\mathbf{v}\cdot\nabla\mathbf{n})\cdot(\nabla\mathbf{n})^T], \quad (15a)$$

$$\Phi=[((\nabla\mathbf{n})\cdot(\nabla\mathbf{n})^T)^{-1}]^T, \quad (15b)$$

where Φ is the anisotropic permeability, and $f_g=\varrho(\mathbf{g}\cdot\mathbf{x})$ is the gravitational energy. This equation is an alternative and generalized expression of the linear momentum balance equation for CLCs in the absence of inertia. In the absence of significant viscous deformation $(\mathbf{A}=\mathbf{0},\mathbf{W}=\mathbf{0})$ the Leslie Eq. (15) generalizes Darcy law

$$\mathbf{v}(\mathbf{x})=\frac{\Phi}{\gamma_1}\cdot[-\nabla(p+f_d+f_m+f_g)]. \quad (16)$$

Projecting Eq. (15) along (∥) and normal (⊥) to the pitch direction $(\mathbf{k})$, we find the velocity fields $(\mathbf{v}_\parallel,\mathbf{v}_\perp)$

$$\mathbf{v}_\parallel(\mathbf{x})=\frac{\Phi}{\gamma_1}:[-\nabla(p+f_d+f_m+f_g)+\nabla\cdot\mathbf{t}^{\mathrm{ve}}-(\mathbf{h}^v-\gamma_1\mathbf{v}\cdot\nabla\mathbf{n})\cdot(\nabla\mathbf{n})^T]\cdot\mathbf{kk}, \quad (17a)$$

$$\mathbf{v}_\perp(\mathbf{x})=\frac{\Phi}{\gamma_1}:[-\nabla(p+f_d+f_m+f_g)+\nabla\cdot\mathbf{t}^{\mathrm{ve}}-(\mathbf{h}^v-\gamma_1\mathbf{v}\cdot\nabla\mathbf{n})\cdot(\nabla\mathbf{n})^T]\cdot(\mathbf{I}-\mathbf{kk}). \quad (17b)$$

Equations (17a), (17b) is the LE linear momentum balance equations along the helix and perpendicular to the helix, respectively, in the absence of inertia, and in the presence of arbitrary director distortions. Next, we focus on the use of Eq. (17a) to analyze slow permeation flow problems when inertia is negligible and director distortions are weak. In the small director distortions regime, several terms in Eq. (17a) can be ignored, as shown in what follows. Cholesteric permeation flow exists when the main flow is along the helix axis and the flow is sufficiently weak such that the director field is planar and chiral

$$\mathbf{n}=\mathbf{n}^o+(\mathbf{p}\times\mathbf{n}^o)\varphi, \quad (18)$$

where $\mathbf{p}$ is the unit vector along the helix (z) axis, and φ is a small twist distortion. In the linear regime, the main flow velocity in the z direction, obtained from Eq. (17a) is, in the absence of magnetic fields, given by

$$\mathbf{v}_z(\mathbf{x})=\mathbf{v}_\parallel\cdot\mathbf{k}=\frac{1}{q_o^2\gamma_1}\cdot[-\nabla(p+f_g)+\nabla\cdot\mathbf{t}^{o^{\mathrm{ve}}}]\cdot\mathbf{k}, \quad (19)$$

where the superscript "o" denotes the linear regime. Averaging the linearized viscoelastic stress term in Eq. (19) over the pitch, it is found

$$\nabla\cdot\mathbf{t}^{o^{\mathrm{ve}}}\cdot\mathbf{k}=\eta_f\nabla_\perp^2, \quad (20a)$$

$$\eta_f=\frac{(\eta_a+\eta_c)}{2}, \quad (20b)$$

$$\eta_a=\tfrac{1}{2}\alpha_4, \quad (20c)$$

$$\eta_c=\tfrac{1}{2}(-\alpha_2+\alpha_4+\alpha_5), \quad (20d)$$

where ⊥ denotes the normal plane to $\mathbf{k}$, η_f is the average of the nematic Miesowicz viscosities η_a and η_b [1]. The general expression of the primary velocity for permeation flow obtained without explicity consideration of the director field is

$$v_z(x,y)=\frac{1}{q_o^2\gamma_1}\cdot\left[-\frac{\partial}{\partial z}(p+f_g)+\eta_f\nabla_\perp^2 v_z\right]. \quad (21)$$

III. APPLICATION: GRAVITY-DRIVEN PERMEATION FLOW

As an application of Eq. (21), we consider the gravity-driven flow over an inclined flat plate, with a given flow rate Q, of a CLC of pitch q_o. Let σ be the angle of the plate with the vertical direction, H be the CLC film thickness along the velocity gradient (x) direction, and z be the flow and helix direction. In contrast to pressure-driven Poisueille permeation flow in a capillary of fixed radius R, the film thickness H in film flow is unknown and a function of Q, and the flow will therefore exhibit a Newtonian regime when $H\approx q_o^{-1}$ and a non-Newtonian regime when $H\gg q_o^{-1}$. The boundary conditions and $v_z(x)$ obtained from Eq. (21) are

$$v_z(x)=\frac{1}{q_o^2\gamma_1}\cdot\left[\varrho g\cos\sigma+\eta_f\frac{d^2v_z}{dx^2}\right], \tag{22a}$$

$$x=0,\ \ v_z=0, \tag{22b}$$

$$x=H,\ \frac{dv_z}{dx}=0, \tag{22c}$$

whose solutions is

$$v_z(x)=\frac{\varrho g\cos\varphi}{\gamma_1 q_o^2}[1-\cosh\beta x+\tanh\beta H\sinh\beta x]. \tag{23}$$

The unknown film thickness H is found by computing the known flow rate

$$Q=\int_0^H v_z(x)dx, \tag{24}$$

which yields the following implicit expression for $H(R)$:

$$R=\beta H-\tanh\beta H, \tag{25a}$$

$$R=\frac{Q\beta\gamma_1 q_o^2}{\varrho g\cos\sigma}, \tag{25b}$$

where R is the ratio of viscous time scale to flow time scale for gravity-driven flow. The Newtonian regime emerges when $R\approx 1$. Expanding tanh βH to second order yields the classical Newtonian film thickness expression and the following scalings:

$$H(Q)=\sqrt[3]{\frac{3\eta_f Q}{\varrho g\cos\sigma}}, \tag{26a}$$

$$H\propto Q^{1/3}, \tag{26b}$$

$$H\neq f(q_o). \tag{26c}$$

The film thickness varies nonlinearly with flow rate and is independent of the pitch. The magnitude of H is of the order of q_o. The non-Newtonian regime arises when $R\gg 1$. In this case, tanh $\beta H\ll\beta H$ and the expression and scalings of H are

$$H(Q)=\frac{Q\gamma_1 q_o^2}{\varrho g\cos\sigma}, \tag{27a}$$

$$H\propto Q, \tag{27b}$$

$$H\propto q_o^2. \tag{27c}$$

H varies linearly with Q, depends on q_o, and $H\gg q_o$. The apparent viscosity $\eta_{\rm app}$ for gravity-driven film flow is given by the shear stress at the wall divided by the shear rate at the wall of a Newtonian fluid flowing with flow rate Q

$$\eta_{\rm app}=\frac{\varrho g\cos\sigma H^3}{3Q}. \tag{28}$$

In the Newtonian regime ($R\approx 1$), the apparent viscosity, found from Eqs. (26a) and (28), is the average of the nematic Miesowicz viscosities

$$\eta_{\rm app}=\eta_f=\frac{(\eta_a+\eta_c)}{2}. \tag{29}$$

In the non-Newtonian regime ($R\gg 1$), the apparent viscosity, found from Eqs. (27a) and (28), is shear thickening

$$\eta_{\rm app}=\frac{\eta_f R^2}{3}=\frac{\gamma_1^3 q_o^6 Q^2}{3(\varrho g\cos\sigma)^2}. \tag{30}$$

In the non-Newtonian regime, the apparent viscosity is several orders of magnitude greater than in the Newtonian regime.

In conclusion, we have derived a new expression for the Leslie-Ericksen linear momentum balance equation and have used it to formulate a general equation for the primary velocity in permeation flow of CLCs. The gravity-driven film permeation flow of a CLC over an inclined plate is analyzed. In this free-boundary problem, the system size depends on the flow rate, whose magnitude gives rise to a Newtonian, thin film low apparent viscosity, slow-flow regime, and a non-Newtonian thick film, high apparent viscosity, fast-flow regime.

ACKNOWLEDGMENT

This work is supported by the Air Force Office of Scientific Research, Mathematical and Space Science Program, under Grant No. F49620-00-1-0341.

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