Field-Coupled Mechanics of Finite Deforming Liquid Crystal Elastomers

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Abstract

Liquid crystal elastomers exhibit a number of fascinating material characteristics which are relevant to applications in information technology, robotics, and biomedical devices. These materials produce field-coupled deformation in response to light, chemicals, electric fields, and heat and therefore provide many opportunities for the design of morphing structure applications. Here, we investigate how finite deformation of an elastomer network couples with microscopic liquid crystal domain structure evolution. It is shown that a broad class of liquid crystal elastomer behavior can be predicted using nonlinear continuum mechanics without the use of explicit phenomenological coupling parameters. The governing equation are presented and coupling to the Cauchy stress is given to illustrate how spontaneous strain occurs from liquid crystals embedded within the elastomer network.

1 Introduction

Liquid crystal elastomers exhibit a number of exciting material characteristics that have useful applications in information technology [1], bio-inspired robotics [2], biomedical devices [3], and nanotechnology [4]. Since the pioneering synthesis development by Finkelmann and colleagues [5], interesting compositions continue to be synthesized. For example, these materials can generate deformation from light [2, 6], heat [7], chemicals [8], and electric fields [9]. Moreover, certain bent-core liquid crystal elastomers have demonstrated flexoelectricity which is exciting for powering and controlling artificial organs.

The extraordinary coupling behavior afforded by these materials is strongly connected to the liquid crystal mesogen (i.e., liquid crystal forming molecule) and its coupling to the host elastomer network. Liquid crystals are well-known for their electro-optic applications [10], but also offer exciting possibilities for artificial muscle applications. Since polymers are one of the most superior materials due to their light weight properties, ease of processibility, and flexibility in molecular design; they offer an excellent medium for synthesizing highly anisotropic and multifunctional liquid crystal elastomers for such applications.

To effectively utilize these materials in devices, a strong understanding of the underlying liquid crystal structure evolution and coupling with the elastomer network is important. Quantifying how these materials respond to mechanical loads, light, heat, chemicals, or electric fields is necessary to facilitate materials design and materials integration into structures and devices. Here, we discuss the role of finite deformation on the evolution of the underlying liquid crystal structure and elastomer deformation.

This subject falls into the category of nonlinear continuum mechanics which has been intensely studied [11, 12], but general modeling frameworks that

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quantify explicit coupling between an elastomer and the underlying liquid crystal structure are still limited. The solid mechanics of liquid crystal elastomers has been described in [13–17], but complexities due to the interactions between an elastomer network and liquid crystals during finite deformation still raises many questions on the underlying material mechanisms governing field-coupled deformation.

In this paper, we summarize the governing equations of field-coupled, finite deformation in liquid crystal elastomers. It is shown that spontaneous deformation and reorientation of the liquid crystal mesogens can be predicted without introducing any explicit phenomenological coupling. Similar effects have been developed for idealized dielectric elastomers which lead to the classic Maxwell stresses (electrostriction) [18, 19]. Extensions of this concept to liquid crystal elastomers is more complex due to the side or main chain liquid crystal attachment to the polymer backbone as well as the spacers connecting these material constituents. Slight variations in the molecular structure can lead to oblate, prolate, or spherical spontaneous deformation [12]. In this article, we show that all three of these cases can be predicted using a Landau free energy of liquid crystals coupled with a hyperelastic constitutive law of an elastomer network.

The outline of the paper is given as follows. A set of balance laws and thermodynamic relations for the liquid crystal elastomer are given in Section 2 in the current, deformed configuration. In Section 3, a phenomenological energy function is introduced and used to quantify fields and stresses that lead to spontaneous deformation. Concluding remarks summarizing the results and outlook on future developments are given in the final section.

2 Governing Equations

The balance laws are first developed by coupling mechanical forces due to stretching an elastomer network with director forces due to nematic phase liquid crystal domain structures. Stress tensors for the elastomer network and liquid crystals are defined and incorporated into the governing equations. A thermodynamic potential is introduced to quantify conjugate variables for the stress components and effective liquid crystal molecular fields.

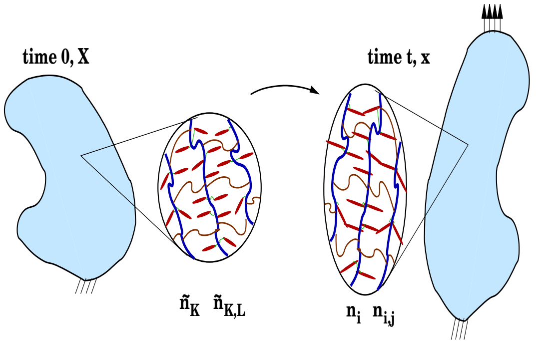

Figure 1: Multiscale material description of a liquid crystal elastomer. Left image: reference (original) state. Right image: current (deformed) state. The director and director gradients are defined in the reference $\tilde{n}_i$ and current n_i configurations.

The nonlinear continuum mechanics model is formulated by defining each material particle of the liquid crystal elastomer in an arbitrary initial reference coordinate denoted by X_K. This follows the description illustrated in Figure 1 where each material point defines the location of the effective liquid crystal elastomer structure. In the current state, a material particle occupies the spatial point defined by $x_i(X_K, t)$ at time t. The stretch of the liquid crystal elastomer is quantified by changes between the spatial point and the reference configuration

$$F_{iK}(X_K, t) = \frac{\partial x_i(X_K, t)}{\partial X_K} \tag{2.1}$$

which is defined as the deformation gradient [20].

Conservation of mass is defined for a material volume

$$\frac{d}{dt}\int_{\Omega} \rho dV = 0 \Rightarrow \frac{d\rho}{dt} + \rho \frac{\partial v_i}{\partial x_i} = 0 \tag{2.2}$$

where $\rho(x_i, t)$ is the mass density, v_i is the velocity of a material point, and Ω is the domain of the material in the current configuration.

Conservation of linear momentum is defined to include contributions from the elastomer and liquid crystal

$$\int_{\Omega} \left(b_i + b_i^L\right) dV + \int_{\Gamma} \left(t_i + t_i^L\right) dS = \frac{d}{dt}\int_{\Omega} \rho v_i dV \tag{2.3}$$

where b_i and b_i^L are body forces due to the elastomer and liquid crystal, respectively. The boundary of the domain in the current configuration is Γ. It is assumed that the body forces due to the liquid crystal can be related to stress by

$$b_i^L = \frac{\partial \sigma_{ji}^L}{\partial x_j} \tag{2.4}$$

Traction is assumed to depend on mechanical effects from the elastomer network (t_i) and the liquid crystals (t_i^L). Linear momentum is satisfied by balancing to the total traction $(t_i + t_i^L)$ with the Cauchy stress, σ_{ij}, by defining

$$t_i + t_i^L = \sigma_{ji}\hat{n}_j \tag{2.5}$$

where the unit normal in the current configuration, not to be confused with the liquid crystal director n_j, is denoted by $\hat{n}_j$. It is assumed that applied mechanical traction on the surface of a volume element is balanced by a combination of internal stresses from the elastomer and the liquid crystals, i.e., $t_i = (\sigma_{ji} + \sigma_{ji}^L)\hat{n}_j$. Whereas stresses within a liquid crystal and an elastomer can be quantified prior to synthesizing a liquid crystal *elastomer*, once the material constituents are chemically bonded, it is assumed that σ_{ij} and σ_{ij}^L may be chemically altered and cannot be individually measured.

Using the above relations, it can be shown that linear momentum is

$$\frac{\partial \sigma_{ji}}{\partial x_j} + \frac{\partial \sigma_{ji}^L}{\partial x_j} + b_i = \rho \frac{dv_i}{dt} \quad \text{in} \quad \Omega \tag{2.6}$$

This equation will be coupled to a set of director forces and conjugate variables that describe liquid crystal structure evolution.

2.1 Liquid Crystal Director Forces

A set of liquid crystal director forces are introduced to quantify its effect on the mechanics of the liquid crystal elastomer. Similar to a configuration force balance proposed by Gurtin [21], a liquid crystal director force balance is defined as a function of the liquid crystal director as

$$\int_\Gamma \xi_{ji}\hat{n}_j dS + \int_\Omega (\pi_i + \gamma_i)\, dV = 0 \tag{2.7}$$

where ξ_{ji} is a micro-stress tensor, π_i is an internal micro-force, and γ_i is an external micro-force.

By application of the divergence theorem, (2.7) is

$$\xi_{ji,j} + \pi_i + \gamma_i = 0 \quad \text{in} \quad \Omega \tag{2.8}$$

The variable π_i is defined to include forces at equilibrium and velocity dependent forces. The velocity dependent term must satisfy certain constraints based on the Clausius-Duhem inequality [20], while equilibrium forces are defined in terms of a thermodynamic potential. The internal force is defined by

$$\pi_i = -\eta_i - \beta_{ij}\dot{n}_j \tag{2.9}$$

where the first term is due to conservative forces. The second term includes a positive definite inverse mobility tensor β_{ij} and a time rate of change of the director, $\dot{n}_j$. These relations along with conjugate variables for the elastomer network are given in the following section.

2.2 Thermodynamic Relations

A Helmholtz energy function per unit current volume is defined by

$$\psi = \psi(F_{iK}, n_i, n_{i,j}, \dot{n}_i) \tag{2.10}$$

where $\dot{n}_i$ is the material time derivative on the director. This term is initially considered in the free energy since $\dot{n}_i$ was included in the definition of π_i.

The free energy relation between the current and reference volume is defined by

$$\Psi = \int_\Omega \psi dV = \int_{\Omega_0} \psi J dV_0 = \int_{\Omega_0} \widetilde{\psi} dV_0 \tag{2.11}$$

where dV is the current volume element over the domain Ω in the current configuration and dV_0 is the reference volume element over the domain Ω_0 in the reference configuration. The relation $dV = JdV_0 = \det(F_{iK})dV_0$ has also been used. These relations provide the transformation

$$\widetilde{\psi} = J\psi \tag{2.12}$$

According to the second law of thermodynamics, the time rate of change of the free energy and kinetic energy must be less than or equal to the externally applied power. Including effects from both the liquid crystal director and elastomer, this is stated as

$$\int_\Omega \dot{\psi} dV \leq \int_\Omega \left[\left(\sigma_{ji} + \sigma_{ji}^L\right) v_{i,j} + \xi_{ji}\dot{n}_{i,j} - \pi_i \dot{n}_i\right] dV \tag{2.13}$$

which is assume to apply to every volume element according to

$$\dot{\psi} \leq \left(\sigma_{ji} + \sigma_{ji}^L\right) v_{i,j} + \xi_{ji}\dot{n}_{i,j} - \pi_i \dot{n}_i \tag{2.14}$$

The time rate of change of the deformation gradient is

$$\frac{dF_{iK}}{dt} = \frac{\partial x_j}{\partial X_K}\frac{\partial v_i}{\partial x_j} \tag{2.15}$$

which is used to obtain the time rate of change of the free energy in (2.14) according to

$$\dot{\psi} = \frac{\partial \psi}{\partial F_{iK}} F_{jK} v_{i,j} + \frac{\partial \psi}{\partial n_i}\dot{n}_i + \frac{\partial \psi}{\partial n_{i,j}}\dot{n}_{i,j} + \frac{\partial \psi}{\partial \dot{n}_i}\ddot{n}_i \tag{2.16}$$

The free energy therefore must be independent of the material time derivative of the director such that $\dfrac{\partial\psi}{\partial\dot{n}_i} = 0 \Rightarrow \psi = \psi(F_{iK}, n_i, n_{i,j})$ by comparison to (2.14); see [22] for similar relations on ferroelectric domain structures.

By comparisons of (2.14) and (2.16), the conjugate variables for the stress and micro-stress tensors are

$$\begin{aligned} \sigma_{ji} + \sigma^L_{ji} &= F_{jK}\frac{\partial\psi}{\partial F_{iK}} \\ \xi_{ji} &= \frac{\partial\psi}{\partial n_{i,j}} \end{aligned} \tag{2.17}$$

where the Cauchy stress relation can also be defined using the energy transformation given by (2.12) which gives the conventional relation $\sigma_{ji} + \sigma^L_{jk} = J^{-1}F_{jK}\dfrac{\partial\widetilde{\psi}}{\partial F_{iK}}$ [20]. The relation $\dfrac{\partial\widetilde{\psi}}{\partial F_{iK}}$ is known as the nominal stress. This relation will be defined in the following section to quantify stresses due to the elastomer network and liquid crystal mesogens in the reference configuration.

A conjugate variable is also defined for the effective liquid crystal field as $\eta_i = \dfrac{\partial\psi}{\partial n_i}$ which is equivalent to the conservative force in π_i previously given in (2.9). This relation requires

$$\left(\eta_i + \pi_i\right)\dot{n}_i \le 0 \tag{2.18}$$

based on (2.14) such that the inverse mobility tensor defined in (2.9) must be positive definite for any arbitrary change in the director according to

$$\beta_{ij}\dot{n}_j\dot{n}_i \ge 0 \tag{2.19}$$

which is consistent with the arguments given in Section 2.1.

3 Energy Relations

The constitutive relations governing the liquid crystal elastomer is formulated by introducing a free energy description that is a function of the director order parameter and deformation gradient. The free energy per unit current volume is divided into the following energy components per unit current volume

$$\psi = \psi_M(F_{iK}) + \psi_L(n_i, n_{i,j}) \tag{3.1}$$

where the director and director gradient $n_{i,j}$ are both defined in the current configuration. It is assumed that the free energy of the liquid crystal material, $\psi_L(n_i, n_{i,j})$, and elastomer network, $\psi_M(F_{iK})$, are decoupled in the current configuration. This avoids potential unnecessary intricacies in the material description; however, certain compositions may deviate from this "idealized" case. Additional coupling terms may be introduced into (3.1) to accommodate discrepancies with experiments; however, the "idealized" case will be considered here to show how certain coupling occurs when finite deformation is employed.

The liquid crystal energy function is further decomposed into the components $\psi_L(n_i, n_{i,j}) = \psi_{La}(n_i) + \psi_{Fr}(n_{i,j})$ to quantify anisotropic director and director gradient effects on the elastomer. The term $\psi_{La}(n_i)$ is the Landau energy describing liquid crystal phase and $\psi_{Fr}(n_{i,j})$ is the Frank elastic energy or disortional energy present in polydomain configurations. Elastic and hyperelastic energy functions describing elastomers are well-known and the formulation given here can be used to incorporate any of these energy functions into $\psi_M(F_{iK})$; see [23] for several examples. A description of the Landau energy and the corresponding stresses are given as follows. Similar results can be obtain for stresses near domain walls. For brevity, polydomain stresses and the elastomer energy function are not described here; see [24] for details on these terms.

The liquid crystal energetics are normally defined in terms of quadrapole effects where the trace of the director magnitude ($\frac{1}{3}n_i n_i$, summation implied) is subtracted out of the energy function. This approach assumes hydrostatic deformation does not contribute to the energy. A traceless second order tensor in terms of the director is thus introduced as

$$Q_{ij} = \frac{Q}{2}\left(3n_i n_j - \delta_{ij}\right) \tag{3.2}$$

Prior to defining the energy and field-coupled stress relations, a set of definitions for the liquid crystal fields and stresses in the reference and spatial configurations are given. The liquid crystal director and Cauchy stress are related in the spatial and reference configurations by

$$\begin{aligned} n_i &= J^{-1}F_{iK}\widetilde{n}_K \\ Q_{ij} &= J^{-1}F_{iK}F_{jL}\widetilde{Q}_{KL} \\ \sigma_{ij} + \sigma^L_{ij} &= J^{-1}F_{jK}\left(s_{iK} + s^L_{iK}\right) \end{aligned} \tag{3.3}$$

where the nominal stress has been defined as $s_{iK} + s^L_{iK} = \dfrac{\partial\widetilde{\psi}}{\partial F_{iK}}$ following (2.12) and $(2.17)_1$. The director and second order director tensor in the reference configuration are $\widetilde{n}_K$ and $\widetilde{Q}_{KL}$, respectively.

The effective molecular field associated with the liquid crystal director is

$$\eta_i = H_{iK}\widetilde{\eta}_K \tag{3.4}$$

where H_{iK} is the inverse deformation gradient which has the properties, $H_{iK}F_{jK} = \delta_{ij}$ and $H_{iK}F_{iL} = \delta_{KL}$ where δ_{ij} and δ_{KL} are the Kronecker deltas [20].

3.1 Landau Liquid Crystal Energy Relations

The Landau energy component for the liquid crystal energy is defined by a polynomial expansion with second, third, and fourth order components on the second order tensor, Q_{ij}; see [12,25] for a similar energy function. In the spatial configuration, the Landau energy is

$$\begin{aligned}\psi_{La}(Q_{ij}) \quad = \quad & \frac{A(T)}{2}Q_{ij}Q_{ji} - \frac{B(T)}{3}Q_{ij}Q_{jk}Q_{ki} + \dots \\ & \frac{C(T)}{4}(Q_{ij}Q_{ij})^2\end{aligned} \tag{3.5}$$

where $A(T)$, $B(T)$, and $C(T)$ are phenomenological constants that define the liquid crystal phase characteristics. These terms are often defined to be strongly temperature (T) dependent. In this analysis, the coefficients are considered under isothermal conditions at temperatures below the isotropic-to-nematic transition point.

The effective molecular field in the spatial configuration is

$$\eta_i = \frac{\partial \psi}{\partial Q_{kl}}\frac{\partial Q_{kl}}{\partial n_i} \tag{3.6}$$

Using (2.12), $(3.3)_1$, and (3.5), the Landau energy can be written in terms of $\widetilde{Q}_{KL}$ in the reference configuration

$$\widetilde{\psi}_{La} = \widetilde{\psi}_{La}(F_{iK}, \widetilde{Q}_{KL}) \tag{3.7}$$

which gives an effective molecular field in the reference configuration

$$\widetilde{\eta}_K = \frac{\partial \widetilde{\psi}}{\partial \widetilde{Q}_{IJ}}\frac{\partial \widetilde{Q}_{IJ}}{\partial \widetilde{n}_K}. \tag{3.8}$$

It can be shown that the effective molecular fields η_i and $\widetilde{\eta}_K$ are consistent in the spatial and reference domains [24].

For brevity, we focus on the field-coupled deformation to illustrate how spontaneous strain of the elastomer network forms from the Landau energy. The mechanical coupling between the liquid crystal director and elastomer network is obtained by determining the nominal stress relation from the energy function in the reference configuration (3.7). The nominal stress based on the total Helmholtz energy function is

$$s_{iK} = \frac{\partial \widetilde{\psi}}{\partial F_{iK}} = \frac{\partial \widetilde{\psi}_M}{\partial F_{iK}} + \frac{\partial \widetilde{\psi}_{La}}{\partial F_{iK}} \tag{3.9}$$

where the first term on the right hand side is stress from stretching the elastomer network and the second term is due to the liquid crystal structure.

For the case of the monodomain, we determine the stresses due to the liquid crystals. For brevity, we do not show the elastic effects of the elastomer, but this can be directly incorporated into the formulation. The nominal stress is a lengthy equation; therefore, we show the liquid crystal Cauchy stress following $(3.3)_3$.

$$\begin{aligned}\sigma_{ij}^{La} = & \, 2A(T)Q_{jk}Q_{ki} - \frac{A(T)}{2}Q_{rs}Q_{rs}\delta_{ij} \dots \\ & -2B(T)Q_{jk}Q_{kl}Q_{li} + \frac{2B(T)}{3}Q_{qr}Q_{rs}Q_{st}\delta_{ij} \dots \\ & +2C(T)Q_{jk}Q_{ik}Q_{lm}Q_{lm} - \frac{3C(T)}{4}Q_{qr}Q_{qr}Q_{qr}Q_{qr}\delta_{ij}\end{aligned} \tag{3.10}$$

A substitution of typical values for $A(T)$, $B(T)$, and $C(T)$ illustrate anisotropic deformation that is proportional to the director orientation [12]. Typically $A(T) < 0$ in the nematic phase. Using such values for $A(T)$ and varying $B(T)$ near its nominal value shows that prolate, oblate, and spherical shape changes can be obtained. Note that this has been obtained without any explicit coupling in the energy function. The result given in (3.10) is a consequence of implementing finite deformation in the energy description and governing equations.

4 Concluding Remarks

The governing equations and a thermodynamic energy function has been introduced to describe a relatively broad class of liquid crystal elastomers. A Landau energy was introduced and implemented in the reference domain to obtain coupling to the elastomer network. This formulation avoids introducing additional explicit coupling parameters in the energy function which simplifies the modeling framework. Moreover, implementation of typical Landau energy parameters and elastic coefficients leads to significantly large spontaneous deformation on the order of 10% to > 100% and therefore should not be neglected when modeling liquid crystal elastomers. The modeling framework is expected to be useful in predicting the constitutive behavior of a broad range of field coupled effects such as photomechanics, electrostriction, and chemically driven shape changes.

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