Modeling the collagen fibril network of biological tissues as a nonlinearly elastic material using a continuous volume fraction distribution function

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Abstract

Despite distinct mechanical functions, biological soft tissues have a common microstructure in which a ground matrix is reinforced by a collagen fibril network. The microstructural properties of the collagen network contribute to continuum mechanical tissue properties that are strongly anisotropic with tensile-compressive asymmetry. In this study, a novel approach based on a continuous distribution of collagen fibril volume fractions is developed to model fibril reinforced soft tissues as nonlinearly elastic and anisotropic material. Compared with other approaches that use a normalized number of fibrils for the definition of the distribution function, this representation is based on a distribution parameter (i.e. volume fraction) that is commonly measured experimentally while also incorporating pre-stress of the collagen fibril network in a tissue natural configuration. After motivating the form of the collagen strain energy function, examples are provided for two volume fraction distribution functions. Consequently, collagen second-Piola Kirchhoff stress and elasticity tensors are derived, first in general form and then specifically for a model that may be used for immature bovine articular cartilage. It is shown that the proposed strain energy is a convex function of the deformation gradient tensor and, thus, is suitable for the formation of a polyconvex tissue strain energy function.

1. Introduction

Biological soft tissues, such as cartilage, meniscus, ligament, skin, annulus and artery, have distinct mechanical behaviors and functions but possess a common microstructure in which a porous saturated ground matrix is reinforced by a collagen fibril network. The microstructural arrangement, primary orientation, and mechanical behavior of collagen fibrils contribute to highly anisotropic, nonhomogeneous, and asymmetric tissue material properties. For example, along the depth of articular cartilage fibrils are oriented in the plane parallel to articular surface in the superficial zone [1] whereas they are arranged randomly in the middle zone [2] and turn vertical in the deep zone [1] where they anchor into subchondral bone [3]. In addition, the molecular nature of collagen fibrils results in elastic properties that possess tensile-compressive asymmetry (i.e. tensile moduli are ∼1-2 orders of magnitude greater than compressive moduli) [4, 5]. The range of finite and multi-dimensional strains that such fibrous tissues may experience in vivo [6] suggests the importance, as well as the challenges, of accurately modeling soft tissue biomechanics.

For soft fibrous tissues with multiple constituents, mathematical distribution functions have represented dispersed and continuous (i.e. non-discrete) fibrils oriented in all directions depending on the type of (and anatomical location in) the tissue under investigation [7, 8]. These types of continuous fibril models have been used recently for articular cartilage [9–11]. Those latter models were based on the general structural theory initially proposed in [8] that considered distinct constituent strain energy functions and which calculated the strain energy of the collagen fibril network based on the response of individual fibrils in tension in different directions and integrated over a unit sphere at a material point.

In a mixture theory approach that uses distinct constituents, the total tissue stress is the sum of apparent constituent stresses and, therefore, apparent stresses are commonly used. Then true constituent stresses (or true material constants) are typically multiplied by constituent volume fractions to obtain apparent stresses (or apparent material constants). One aim of this work is to define the relationship between true and apparent collagen network stresses when using a fibril distribution function with fibrils that are mechanically active in tension only.

Thus, the goal of the current study is to develop an accurate nonlinearly elastic and anisotropic model for fibril-reinforced soft tissues. This approach uses true moduli of collagen fibrils multiplied by different volume fractions for different directions to represent the anisotropic distribution of fibrils. Compared with other approaches that use a normalized number of fibrils for the definition of the distribution function, this representation is based on a distribution parameter (i.e. volume fraction) that is commonly measured experimentally. Consequently, an immediate and explicit output of numerical simulation may be the effective volume fraction of the collagen network, i.e. the volume fraction of only those fibrils in tension.

The specific aims are to: 1. introduce the concepts of a continuous volume fraction distribution function and effective volume fraction of the collagen network and derive the corresponding collagen network strain energy function; 2. account for a collagen network that may be "pre-stressed" in a tissue natural configuration; 3. present examples for distribution functions and a specific fibril strain energy function; 4. show that the specific strain energy function is a convex function of the deformation gradient tensor.

2. Preliminaries

It is assumed that the tissue's solid matrix occupies a stress-free reference configuration κ_0 corresponding to a continuous open set of material points in three-dimensional Euclidean space. The tissue is in equilibrium at κ_0 and, under an overall solid matrix deformation gradient tensor $\mathbf{F}$, will occupy the current configuration κ (Figure 1). In biological tissues, the solid matrix is a multiphasic material; e.g., for articular cartilage it can be considered as a mixture of collagens, glycosaminoglycans, and other ground matrix constituents. These constituents may have distinct reference configurations which may or may not be stress-free [12]. Here, focus is on the collagen constituent only.[1]

Due to swelling pressure of glycosaminoglycans, the collagen network is not stress-free at the solid matrix reference configuration. The collagen initial deformation gradient tensor $\mathbf{F}_0^{\mathrm{COL}}$ maps a stress-free collagen reference configuration κ_0^{COL} to the solid matrix reference configuration κ_0. The collagen deformation gradient tensor $\mathbf{F}^{\mathrm{COL}}$ that maps κ_0^{COL} to the current configuration κ is obtained via the multiplicative decompostion

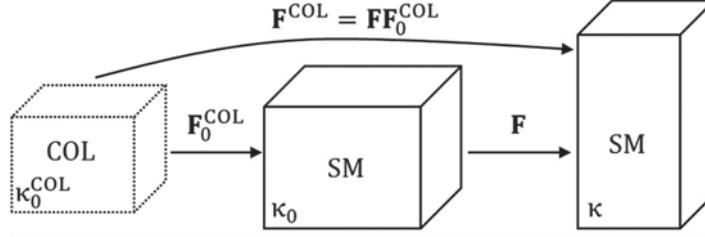$$\mathbf{F}^{\mathrm{COL}} = \mathbf{F} \ \mathbf{F}_0^{\mathrm{COL}} \tag{1}$$

Figure 1. The tissue's solid matrix occupies a stress-free reference configuration κ_0, where constituent pre-stresses balance each other. The deformation gradient tensor $\mathbf{F}_0^{\text{COL}}$ maps the collagen stress-free initial configuration κ_0^{COL} to κ_0.

Consequently, the collagen and solid matrix right Cauchy-Green deformation tensors are related by

$$\mathbf{C}^{\text{COL}} = (\mathbf{F}^{\text{COL}})^{\text{T}}\mathbf{F}^{\text{COL}} = (\mathbf{F}_0^{\text{COL}})^{\text{T}}\mathbf{C}\ \mathbf{F}_0^{\text{COL}} \tag{2}$$

The collagen Lagrangian strain tensor $\mathbf{E}^{\text{COL}}$ defined with respect to the collagen initial configuration κ_0^{COL}, using equation (2), is

$$\mathbf{E}^{\text{COL}} = \frac{1}{2}(\mathbf{C}^{\text{COL}} - \mathbf{I}) = \frac{1}{2}\left((\mathbf{F}_0^{\text{COL}})^{\text{T}}\mathbf{C}\ \mathbf{F}_0^{\text{COL}} - \mathbf{I}\right) \tag{3}$$

Assuming collagen as a nonlinear Green-elastic material, its second Piola-Kirchhoff stress tensor $\mathbf{S}^{\text{COL}}$ is related to its strain energy function W^{COL} as:[2]

$$\mathbf{S}^{\text{COL}} = 2\frac{\partial \text{W}^{\text{COL}}}{\partial \mathbf{C}} \tag{4}$$

Furthermore, the collagen elasticity tensor $\mathbb{C}^{\text{COL}}$ is obtained from W^{COL} via

$$\mathbb{C}^{\text{COL}} = 4\frac{\partial^2 \text{W}^{\text{COL}}}{\partial \mathbf{C}\partial \mathbf{C}} \tag{5}$$

Note that the derivatives in (4) and (5) are defined with respect to $\mathbf{C}$ and, hence, the solid matrix reference configuration κ_0. Thus, when $\mathbf{C} = \mathbf{I}, \mathbf{S}^{\text{COL}} \neq \mathbf{0}$ and indicates the collagen stress in κ_0.

3. Collagen volume fraction distribution and strain energy functions

3.1. Theory

To include the possible contributions of collagen fibrils in all directions, a local spherical coordinate system at a material point is used [7, 9]. At a material point, fibrils inside a pyramidal volume element dV cross through the differential area $\text{dA} = \sin\Theta\text{d}\Theta\text{d}\Phi$ with outward normal $\mathbf{n}$ (Fig. 2). The apex of a pyramidal volume element dV is located at the center of the sphere and its base is the surface element dA. Thus, the differential volume of a pyramidal element is

$$\text{dV} = \frac{1}{3}\sin\Theta\ \text{d}\Theta\text{d}\Phi \tag{6}$$

Fibrils inside a pyramidal volume element are oriented in the range $[(\Theta, \Theta + \text{d}\Theta), (\Phi, \Phi + \text{d}\Phi)]$ with $0 \leq \Theta \leq \pi$ and $0 \leq \Phi \leq 2\pi$.

Here, a *volume fraction* distribution function is used to define the proportion of collagen fibrils oriented in different directions, so that the *total volume fraction* defined as the integral of the volume fraction distribution function over the unit sphere equals the volume fraction of fibrils at a material point in the tissue. Specifically, this distribution function assigns a certain percentage of the total volume fraction of collagen to dV identified by an average outward normal vector $\mathbf{n}$. Similarly, the total strain energy function will be the integral of the
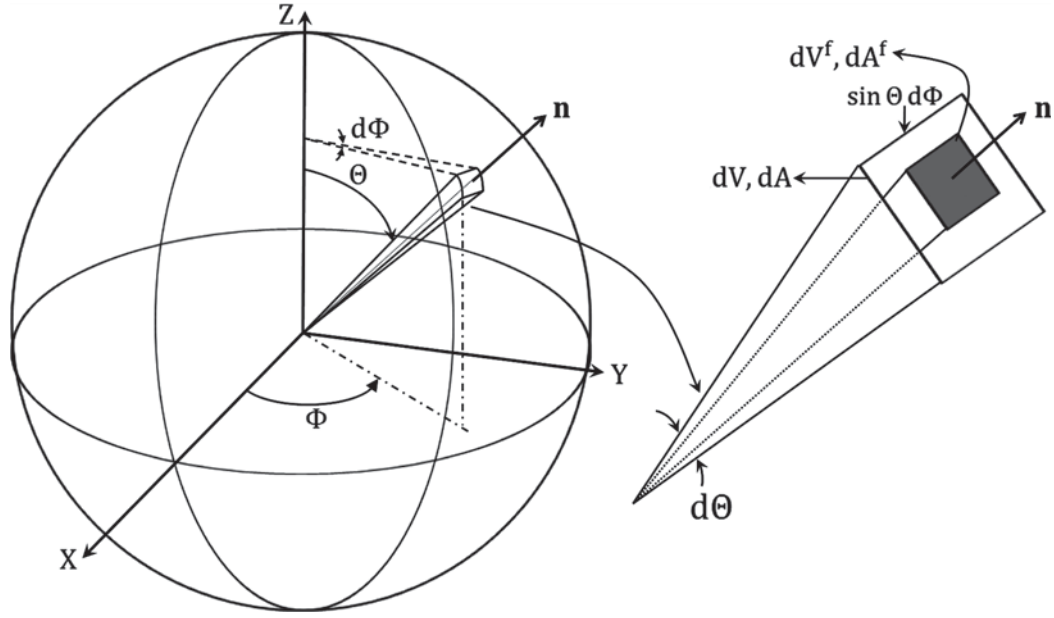
Figure 2. Representation of the unit sphere and a typical pyramidal volume element dV with its corresponding area base dA. The fibrils can be represented by a smaller pyramidal volume element dV^f with area base dA^f (in grey).

uniaxial fibril strain energy function in each direction $\mathbf{n}$ over the unit sphere. $E_\mathbf{n}^{COL}$, the fibril strain in the direction $\mathbf{n}$, is related to the collagen Lagrangian strain tensor $\mathbf{E}^{COL}$ via

$$E_\mathbf{n}^{COL} = \mathbf{n} \cdot [\mathbf{E}^{COL}\mathbf{n}] \tag{7}$$

where $(\cdot)$ is the dot product. The Heaviside step function $H(E_\mathbf{n}^{COL})$ is used so that fibrils in compression do not contribute to the strain energy function i.e. $H(E_\mathbf{n}^{COL}) = 1$ or 0 if $E_\mathbf{n}^{COL} \geq 0$ or < 0, respectively. The total volume fraction of fibrils (defined as $\phi_{tot}^f = \dfrac{V^f}{V^{tot}}$ where V^f and V^{tot} are the fibril volume and unit sphere volume, respectively) will be modified by $H(E_\mathbf{n}^{COL})$ to become an *effective volume fraction*. Thus, the effective volume fraction is the volume fraction of fibrils in tension only and, consequently, depends on the strain at a material point.

In order to motivate the strain energy definition, first consider the special case where the collagen fibrils with the true stain energy density function Ψ, with units of energy per volume, are oriented isotropically and occupy the entire volume of the unit sphere at a material point i.e. $V^f = V^{tot}$. The collagen strain energy density, with units of energy per volume, is

$$W^{COL} = \frac{1}{V^{tot}} \int_V H(E_\mathbf{n}^{COL})\Psi(E_\mathbf{n}^{COL})dV \tag{8}$$

However, in a biological tissue there exist multiple constituents at a typical material point and the isotropically distributed collagen fibrils occupy a volume fraction ϕ_{tot}^f. Due to the assumed isotropic distribution, the directional volume fraction of collagen fibrils per pyramidal volume element dV oriented in direction $\mathbf{n}$ and denoted by $\phi_\mathbf{n}^f$ is equal to total volume fraction ϕ_{tot}^f in the unit sphere, i.e.

$$\phi_\mathbf{n}^f = \frac{dV^f}{dV} = \frac{V^f}{V^{tot}} = \phi_{tot}^f \tag{9}$$

Consequently, the collagen strain energy density at a typical material point is[3]

$$W^{COL} = \frac{1}{V^{tot}} \int_V \phi_\mathbf{n}^f H(E_\mathbf{n}^{COL})\Psi(E_\mathbf{n}^{COL})dV \tag{10}$$

Having considered the special case above, now consider an anisotropic distribution of collagen fibrils. The *directional volume fraction per sphere volume* $d\bar{\phi}_{\mathbf{n}}^{f}$ (as opposed to *directional volume fraction per pyramidal volume* $\phi_{\mathbf{n}}^{f}$) is defined as

$$d\bar{\phi}_{\mathbf{n}}^{f} = \frac{dV^f}{V^{tot}} \tag{11}$$

Using (9) and (11), $\phi_{\mathbf{n}}^{f}$ and $d\bar{\phi}_{\mathbf{n}}^{f}$ are related by

$$d\bar{\phi}_{\mathbf{n}}^{f} = \frac{dV}{V^{tot}}\phi_{\mathbf{n}}^{f} \tag{12}$$

Note that $d\bar{\phi}_{\mathbf{n}}^{f} \ll \phi_{\mathbf{n}}^{f}$.

The fibril volume fraction distribution function $R(\theta, \phi)$ is defined for dV spanning $[[\Theta, \Theta + d\Theta], [\Phi, \Phi + d\Phi]]$ as

$$R(\theta, \phi)dV = \frac{dV^f}{V^{tot}} = d\bar{\phi}_{\mathbf{n}}^{f} \tag{13}$$

Using (9) and (12) in (13), one obtains

$$R(\theta, \phi) = \frac{\phi_{\mathbf{n}}^{f}}{V^{tot}} \tag{14}$$

Based on the above definition, the volume fraction distribution function must satisfy

$$\int_{V} R(\theta, \phi)dV = \int_{V} d\bar{\phi}_{\mathbf{n}}^{f} = \phi_{tot}^{f} \tag{15}$$

This definition is different from those in [7,10] where the distribution function must satisfy $\int_{A} R(\theta, \phi)dA = 1$ or $\frac{1}{4\pi} \int_{A} R(\theta, \phi)dA = 1$.

Using (6) and (13), the collagen network strain energy density in (10) becomes

$$W^{COL} = \frac{1}{3} \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} R(\theta, \phi)H(E_{\mathbf{n}}^{COL})\Psi(E_{\mathbf{n}}^{COL})\sin\theta d\theta d\phi \tag{16}$$

3.2. Examples

For further clarification, two examples for distribution function (one discrete, one continuous) are presented.

Example 1. Consider a unit sphere divided into 20 identical pyramidal volume elements dV. Because the total volume of the sphere is $V^{tot} = \frac{4\pi}{3}$, the volume of each $dV = 0.05V^{tot}$.

Now, assume that there exist collagen fibrils with 9% volume fraction (normalized by the total volume of the sphere i.e. $\phi_{tot}^{f} = 0.09V^{tot}$) but that these fibrils exist in only three pyramidal elements. Represent these three pyramidal elements by numbers 1, 2 and 3 and assume that from the 9% collagen volume fraction, 3% exists in pyramid 1 (i.e. $0.03V^{tot}$), 4% exists in pyramid 2 (i.e. $0.04V^{tot}$), and 2% exists in pyramid 3 (i.e. $0.02V^{tot}$). The directional volume fractions per pyramidal volume are

$$\begin{aligned}
\phi_{\mathbf{n}_1}^{f} &= \frac{dV^{f_1}}{dV^1} = \frac{0.03V^{tot}}{0.05V^{tot}} = 0.6 \\
\phi_{\mathbf{n}_2}^{f} &= \frac{dV^{f_2}}{dV^2} = \frac{0.04V^{tot}}{0.05V^{tot}} = 0.8 \\
\phi_{\mathbf{n}_3}^{f} &= \frac{dV^{f_3}}{dV^3} = \frac{0.02V^{tot}}{0.05V^{tot}} = 0.4
\end{aligned} \tag{17}$$

These values show that 60%, 80% and 40% of each pyramidal volume, respectively, is occupied by collagen fibrils. In the remaining 17 pyramids $\phi_{\mathbf{n}_i}^f = 0$ (for i = 4 to 20).

Now, the infinitesimal directional volume fractions per sphere volume for each pyramidal volume element are

$$d\bar{\phi}_{\mathbf{n}_1}^{f_1} = \frac{0.03 V^{tot}}{V^{tot}} = 0.03$$

$$d\bar{\phi}_{\mathbf{n}_2}^{f_2} = \frac{0.04 V^{tot}}{V^{tot}} = 0.04 \tag{18}$$

$$d\bar{\phi}_{\mathbf{n}_3}^{f_3} = \frac{0.02 V^{tot}}{V^{tot}} = 0.02$$

and $d\bar{\phi}_{\mathbf{n}_i}^f = 0$ (for i = 4 to 20). Since $d\bar{\phi}_{\mathbf{n}}^f$ is defined by normalizing the fibril volume inside a pyramidal element to the total sphere volume, $\sum_{i=1}^{20} d\bar{\phi}_{\mathbf{n}_i}^f = \phi_{tot}^f$. On the contrary, since $\phi_{\mathbf{n}_i}^f$ is defined by normalizing the fibril volume inside a pyramidal element to the pyramidal element volume, $\sum_{i=1}^{20} d\phi_{\mathbf{n}_i}^f = \phi_{tot}^f$.

The volume fraction distribution function must satisfy (15)

$$\sum_{i=1}^{20} R(\theta_i, \phi_i) dV = \phi_{tot}^f \tag{19}$$

For this example

$$\sum_{i=1}^{3} R(\theta_i, \phi_i) dV = (R_1(\theta_1, \phi_1) + R_2(\theta_2, \phi_2) + R_3(\theta_3, \phi_3)) dV = \phi_{tot}^f \tag{20}$$

Thus, the corresponding distribution function will be defined as

$$R(\theta_i, \phi_i) = \begin{cases} (0.03/(0.05 V^{tot})) & \mathbf{n}_1 \text{ or } (\theta_1, \phi_1) \\ 0.04/(0.05 V^{tot}) & \mathbf{n}_2 \text{ or } (\theta_2, \phi_2) \\ 0.02/(0.05 V^{tot}) & \text{for} \quad \mathbf{n}_3 \text{ or } (\theta_3, \phi_3) \\ 0.0 & \mathbf{n}_i \text{ or } (\theta_i, \phi_i), i = 4 \text{ to } 20 \end{cases} \tag{21}$$

For the special case where all fibrils are in tension, $H(E_{\mathbf{n}}^{COL})$ in all directions and one obtains

$$W^{COL} = \sum_{i=1}^{3} R(\theta_i, \phi_i) \Psi dV = \Psi \left(\frac{0.03}{dV} + \frac{0.04}{dV} + \frac{0.02}{dV} \right) dV = 0.09\Psi \tag{22}$$

Example 2. To define isotropically distributed fibrils with a constant value for all directions (e.g. for the transitional middle zone of articular cartilage) one divides the total volume fraction of collagen ϕ_{tot}^f by the total volume of the sphere to obtain

$$R(\theta, \phi) = \phi_{tot}^f \frac{3}{4\pi} \tag{23}$$

For a 9% total volume fraction of fibrils that are isotropically distributed the distribution function is $R(\theta, \phi) = 0.09 \frac{3}{4\pi}$. For this special case, the integral in (15) becomes

$$\phi_{tot}^f = \frac{1}{3} \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} \left(0.09 \frac{3}{4\pi} \right) \sin\theta d\theta d\phi = 0.09 \tag{24}$$

If all fibrils are in tension, equation (16) takes the specific form

$$W^{COL} = \frac{1}{3} \int_{\theta=0}^{\pi} \int_{\phi=0}^{2\pi} \left(0.09 \frac{3}{4\pi} \right) \Psi(E_{\mathbf{n}}^{COL}) \sin\theta d\theta d\phi = 0.09\Psi \tag{25}$$

4. Collagen network stress and elasticity tensors

Considering $\mathbf{F}_0^{\mathrm{COL}}$ fixed, $\mathrm{E}_\mathbf{n}^{\mathrm{COL}}$ is a function of $\mathbf{C}$ and $\mathbf{n}$; using equations (3) and (7) one obtains

$$E_\mathbf{n}^{\mathrm{COL}} = \tilde{E}_\mathbf{n}^{\mathrm{COL}}(\mathbf{C}, \mathbf{n}) = \mathbf{n} \cdot \left[\left[\frac{1}{2}\ (\mathbf{F}_0^{\mathrm{COL}})^{\mathrm{T}} \mathbf{C}\, \mathbf{F}_0^{\mathrm{COL}} - \mathbf{I}\ \right] \mathbf{n}\right] \tag{26}$$

Thus, equation (16) can be written in terms of $\mathbf{C}$ as

$$W^{\mathrm{COL}} = \int_V R(\theta, \phi)\tilde{H}(\mathbf{C}, \mathbf{n})\tilde{\Psi}(\mathbf{C}, \mathbf{n})dV \tag{27}$$

Recalling (4) and (5), the collagen stress and elasticity tensors become

$$\mathbf{S}^{\mathrm{COL}} = 2 \int_V R(\theta, \phi)\tilde{H}(\mathbf{C}, \mathbf{n})\frac{\partial}{\partial \mathbf{C}}[\tilde{\Psi}(\mathbf{C}, \mathbf{n})]dV \tag{28}$$

$$\tilde{\mathbb{C}}^{\mathrm{COL}} = 4 \int_V R(\theta, \phi)\tilde{H}(\mathbf{C}, \mathbf{n})\frac{\partial^2}{\partial \mathbf{C}\partial \mathbf{C}}[\tilde{\Psi}(\mathbf{C}, \mathbf{n})]dV \tag{29}$$

4.1. Example

This example illustrates how stress and elasticity tensors can be specified using equations (28) and (29). A quadratic strain energy function may be reasonable for immature native or engineered tissues which do not exhibit strong nonlinearity at large deformations; e.g. immature native tissue has been shown not to exhibit a strong nonlinear response in tension up to 20% strain and compression up to 45% strain [13, 14] so a quadratic strain energy function may be sufficient for some studies. Consider the special form for the true fibril strain energy function

$$\Psi = \Psi(E_\mathbf{n}^{\mathrm{COL}}) = \frac{1}{2}E^{\mathrm{f}}(E_\mathbf{n}^{\mathrm{COL}})^2 \tag{30}$$

where E^{f} is the true collagen elastic modulus.

Using (26) the collagen strain energy density (30) becomes

$$\Psi = \frac{1}{2}E^{\mathrm{f}}\ \left(\mathbf{n} \cdot \left[\left[\frac{1}{2}\ (\mathbf{F}_0^{\mathrm{COL}})^{\mathrm{T}} \mathbf{C}\, \mathbf{F}_0^{\mathrm{COL}} - \mathbf{I}\ \right] \mathbf{n}\right]\right)^2 \tag{31}$$

4.1.1. Stress To calculate $\mathbf{S}^{\mathrm{COL}}$ using equations (28) and (31), recall that the directional derivative of any scalar-valued function $f(\mathbf{Y})$ of a second order tensor $\mathbf{Y}$ with respect to $\mathbf{Y}$ in the direction of an arbitrary second-order tensor $\mathbf{Z}$ is defined as

$$\frac{\partial f}{\partial \mathbf{Y}} : \mathbf{Z} = \left[\frac{\mathrm{d}}{\mathrm{d}\alpha}f(\mathbf{Y} + \alpha\mathbf{Z})\right]_{\alpha=0} \tag{32}$$

where (:) is the scalar double dot product. Replacing $\mathbf{Y}$ with $\mathbf{C}$ and $f(\mathbf{Y})$ with $\tilde{\Psi}(\mathbf{C}, \mathbf{n})$ leads to

$$\frac{\partial \Psi}{\partial \mathbf{C}} : \mathbf{Z} = \left[\frac{\mathrm{d}}{\mathrm{d}\alpha}\tilde{\Psi}(\mathbf{C} + \alpha\mathbf{Z}, \mathbf{n})\right]_{\alpha=0} = \left[\frac{\mathrm{d}}{\mathrm{d}\alpha}\left(\frac{1}{2}E^{\mathrm{f}}\ \left(\mathbf{n} \cdot \left[\left[\frac{1}{2}\ (\mathbf{F}_0^{\mathrm{COL}})^{\mathrm{T}}[\mathbf{C} + \alpha\mathbf{Z}]\mathbf{F}_0^{\mathrm{COL}} - \mathbf{I}\ \right] \mathbf{n}\right]\right)^2\right)\right]_{\alpha=0} \tag{33}$$

After some straightforward tensor algebraic manipulation, one obtains

$$\frac{\partial \Psi}{\partial \mathbf{C}} : \mathbf{Z} = \frac{E^{\mathrm{f}}}{4}\left[\mathbf{n} \cdot \left[\left[(\mathbf{F}_0^{\mathrm{COL}})^{\mathrm{T}} \mathbf{C}\, \mathbf{F}_0^{\mathrm{COL}} - \mathbf{I}\right]\mathbf{n}\right]\right]\left[\mathbf{n} \cdot \left[\left[(\mathbf{F}_0^{\mathrm{COL}})^{\mathrm{T}}\mathbf{Z}\, \mathbf{F}_0^{\mathrm{COL}}\right]\mathbf{n}\right]\right] \tag{34}$$

With further manipulation of the right hand side of (34) and since $\mathbf{Z}$ is arbitrary, the collagen network stress tensor $\mathbf{S}^{\text{COL}}$ in (28) becomes

$$\mathbf{S}^{\text{COL}} = \frac{1}{4} \int_V R(\theta, \phi) \tilde{H}(\mathbf{C}, \mathbf{n}) E^f \left\{ \mathbf{n} \cdot \left[(\mathbf{F}_0^{\text{COL}})^T \mathbf{C} \mathbf{F}_0^{\text{COL}} - \mathbf{I} \right] \mathbf{n} \left[[\mathbf{n} \otimes \mathbf{n}] : (\mathbf{F}_0^{\text{COL}})^T \otimes (\mathbf{F}_0^{\text{COL}})^T \right] \right. $$
$$\left. + \left[\mathbf{F}_0^{\text{COL}} \otimes \mathbf{F}_0^{\text{COL}} \right] : [\mathbf{n} \otimes \mathbf{n}] \right\} dV \tag{35}$$

where $(\otimes)$ denotes the tensor product.

4.1.2. Material elasticity stiffness To calculate $\tilde{\mathbb{C}}^{\text{COL}}$ using equations (29) and (31), the collagen material elasticity tensor is calculated from

$$\tilde{\mathbb{C}}^{\text{COL}} = \frac{1}{2} \int_V R(\theta, \phi) \tilde{H}(\mathbf{C}, \mathbf{n}) E^f \frac{\partial}{\partial \mathbf{C}} \left\{ \mathbf{n} \cdot \left[(\mathbf{F}_0^{\text{COL}})^T \mathbf{C} \mathbf{F}_0^{\text{COL}} - \mathbf{I} \right] \mathbf{n} \left[[\mathbf{n} \otimes \mathbf{n}] : (\mathbf{F}_0^{\text{COL}})^T \otimes (\mathbf{F}_0^{\text{COL}})^T \right] \right. $$
$$\left. + \left[\mathbf{F}_0^{\text{COL}} \otimes \mathbf{F}_0^{\text{COL}} \right] : [\mathbf{n} \otimes \mathbf{n}] \right\} dV \tag{36}$$

Using the directional derivative of a second-order tensor function (i.e. the standard generalization of equation (32)), one obtains

$$\frac{\partial \mathbf{S}}{\partial \mathbf{C}} : \mathbf{Z} = \frac{d}{d\alpha} \left\{ \mathbf{n} \cdot [(\mathbf{F}_0^{\text{COL}})^T (\mathbf{C} + \alpha \mathbf{Z}) \mathbf{F}_0^{\text{COL}} - \mathbf{I}] \mathbf{n} \left[[\mathbf{n} \otimes \mathbf{n}] : (\mathbf{F}_0^{\text{COL}})^T \otimes (\mathbf{f}_0^{\text{COL}})^T \right] \right. $$
$$\left. + \left[\mathbf{F}_0^{\text{COL}} \otimes \mathbf{F}_0^{\text{COL}} \right] : [\mathbf{n} \otimes \mathbf{n}] \right\}_{\alpha=0} \tag{37}$$

After some straightforward manipulation of the right hand side of (37) and using (36), the collagen elasticity tensor is

$$\tilde{\mathbb{C}}^{\text{COL}} = \frac{1}{4} \int_V R(\theta, \phi) \tilde{H}(\mathbf{C}, \mathbf{n}) E^f \left\{ \left[[\mathbf{n} \otimes \mathbf{n}] : (\mathbf{F}_0^{\text{COL}})^T \otimes (\mathbf{F}_0^{\text{COL}})^T + \mathbf{F}_0^{\text{COL}} \otimes \mathbf{F}_0^{\text{COL}} : [\mathbf{n} \otimes \mathbf{n}] \right] \otimes \right.$$
$$\left. \left[[\mathbf{n} \otimes \mathbf{n}] : (\mathbf{F}_0^{\text{COL}})^T \otimes (\mathbf{F}_0^{\text{COL}})^T + [\mathbf{F}_0^{\text{COL}} \otimes \mathbf{F}_0^{\text{COL}}] : [\mathbf{n} \otimes \mathbf{n}] \right] \right\} dV \tag{38}$$

5. Material stability

To address material stability criteria and avoid numerical divergence in computational solutions, a restriction on the strain energy function may be imposed. Here, focus is on the polyconvexity condition[4] for the tissue's solid matrix strain energy function W. A sufficient condition for polyconvexity [21] is as follows: if the strain energy function $W(\mathbf{F})$ satisfies the additive decomposition

$$W(\mathbf{F}) = W_1(\mathbf{F}) + W_2(\det(\mathbf{F})) + W_3(\text{adj}(\mathbf{F})) \tag{39}$$

and each of the functions $W_1(\mathbf{F})$, $W_2(\det(\mathbf{F}))$ and $W_3(\text{adj}(\mathbf{F}))$ is a convex function of, respectively, $\mathbf{F}$, $\det(\mathbf{F})$ and $\text{adj}(\mathbf{f})$, then $W(\mathbf{F})$ is polyconvex. Furthermore, addition of two or more polyconvex functions results in a polyconvex function. For use in tissue models where additional constituent strain energies are added to W^{COL}, it suffices here to prove that W^{COL}, which contributes to the $W_1(\mathbf{F})$ term, is a convex function of $\mathbf{F}$, i.e.:

$$\frac{\partial^2 W^{\text{COL}}}{\partial F_{iP} \partial F_{jQ}} Z_{iP} Z_{jQ} \geq 0 \; \textit{for any arbitrary tensor } Z_{kC} \textit{ with } Z_{kC} = F_{kC} \textit{ and } Z_{kC} = 0_{kC} \tag{40}$$

Considering equation (16), it will be sufficient to show that $\Psi(E_{\mathbf{n}}^{\text{COL}})$ is a convex function of $\mathbf{F}$ because the integrand in equation (16) is a continuous function and, apart from $\Psi(E_{\mathbf{n}}^{\text{COL}})$, the other components of the integrand do not depend on $\mathbf{F}$ and are positive. Therefore, it suffices to show that for $E_{\mathbf{n}}^{\text{COL}} \geq 0$

$$\frac{\partial^2 \Psi(E_{\mathbf{n}}^{\text{COL}})}{\partial F_{iP} \partial F_{jQ}} Z_{iP} Z_{jQ} \geq 0 \; \textit{for any arbitrary tensor } Z_{kC} \textit{ with } Z_{kC} = F_{kC} \textit{ and } Z_{kC} = 0_{kC} \tag{41}$$

C^{COL} in equation (2) and Ψ in equation (31) can be expressed as functions of $\mathbf{F}$

$$C_{\widehat{A}\widehat{B}}^{COL} = F_{m\widehat{A}}^{COL} F_{m\widehat{B}}^{COL} = (F_0^{COL})_{G\widehat{A}} F_{mG} F_{mH} (F_0^{COL})_{H\widehat{B}} \tag{42}$$

$$\Psi = \frac{1}{2} E^f \left[\frac{1}{2} n_{\widehat{A}} n_{\widehat{B}} \left((F_0^{COL})_{G\widehat{A}} F_{mG} F_{mH} (F_0^{COL})_{H\widehat{B}} - \delta_{\widehat{A}\widehat{B}} \right) \right]^2 \tag{43}$$

Noting that the collagen initial configuration is different from the solid matrix reference configuration, the hat superposed on uppercase indices denotes the collagen initial configuration. Calculating the derivative of equation (43) with respect to $\mathbf{F}$ results in

$$\frac{\partial \Psi}{\partial F_{iP}} = \frac{1}{2} E^f \left[n_{\widehat{A}} n_{\widehat{B}} ((F_0^{COL})_{G\widehat{A}} F_{mG} F_{mH} (F_0^{COL})_{H\widehat{B}} - \delta_{\widehat{A}\widehat{B}}) \right] \left[n_{\widehat{K}} n_{\widehat{L}} (F_0^{COL})_{P\widehat{K}} F_{iD} (F_0^{COL})_{D\widehat{L}} \right] \tag{44}$$

Calculating the second derivative of Ψ with respect to $\mathbf{F}$, and with some manipulations, results in

$$\frac{\partial^2 \Psi}{\partial F_{iP} \partial F_{jQ}} = \frac{1}{2} E^f [2 [n_{\widehat{A}} n_{\widehat{B}} (F_0^{COL})_{Q\widehat{A}} F_{jH} (F_0^{COL})_{H\widehat{B}}] [n_{\widehat{K}} n_{\widehat{L}} (F_0^{COL})_{P\widehat{K}} F_{iD} (F_0^{COL})_{D\widehat{L}}]$$
$$+ [n_{\widehat{A}} n_{\widehat{B}} ((F_0^{COL})_{G\widehat{A}} F_{mH} (F_0^{COL})_{H\widehat{B}} - \delta_{\widehat{A}\widehat{B}})] [n_{\widehat{K}} n_{\widehat{L}} (F_0^{COL})_{P\widehat{K}} \delta_{ij} (F_0^{COL})_{D\widehat{L}}]] \tag{45}$$

Recalling (41), (45) becomes

$$\frac{\partial^2 \Psi}{\partial F_{iP} \partial F_{jQ}} Z_{iP} Z_{jQ} = E^f \left[n_{\widehat{A}} Z_{jQ} (F_0^{COL})_{Q\widehat{A}} F_{jH} (F_0^{COL})_{H\widehat{B}} n_{\widehat{B}} \right]^2 + (E_{\mathbf{n}}^{COL}) [Z_{iP} (F_0^{COL})_{P\widehat{K}} n_{\widehat{K}}]^2 \tag{46}$$

A necessary and sufficient condition for the above result to be non-negative is that E^f be positive. Since E^f is always taken positive, the convexity condition is satisfied. It is emphasized that the proof is only valid for the strain energy proposed in equation (30). Note that, for the complete solid matrix strain energy to be polyconvex, the strain energy terms for other constituents need to be considered.

Notes

1. The superscript COL will be used to indicate the collagen constituent.

2. Or in indicial form and since $\mathbf{C}$ is symmetric: $S_{AB}^{COL} = \dfrac{\partial W^{COL}}{\partial C_{AB}} + \dfrac{\partial W^{COL}}{\partial C_{BA}}$.

3. Although $\phi_{\mathbf{n}}^f$ is independent of direction for an isotropic distribution, one cannot factor $\phi_{\mathbf{n}}^f$ out of the integrand in (10) since $H(E_{\mathbf{n}}^{COL})$ must exclude fibrils in compression and generate an effective volume fraction that modulates the true collagen material properties.

4. Polyconvexity guarantees the existence of local minimizers of the strain energy function subject to boundary conditions [16] while not sharing the limitations of convexity related to global uniqueness [17] or invariance requirements and coercivity [18]; see also [15].

Conflict of interest

None declared.

Dedication

This work is dedicated to Professor M. M. Carroll on the occasion of his 75th birthday.

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