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ORIGINAL PAPER

A three-dimensional constitutive model for the stress relaxation of articular ligaments

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Abstract A new nonlinear constitutive model for the three-dimensional stress relaxation of articular ligaments is proposed. The model accounts for finite strains, anisotropy, and strain-dependent stress relaxation behavior exhibited by these ligaments. The model parameters are identified using published uniaxial stress–stretch and stress relaxation data on human medial collateral ligaments (MCLs) subjected to tensile tests in the fiber and transverse to the fiber directions (Quapp and Weiss in J Biomech Eng Trans ASME 120:757–763, 1998; Bonifasi-Lista et al. in J Orthop Res 23(1):67–76, 2005). The constitutive equation is then used to *predict* the nonlinear elastic and stress relaxation response of ligaments subjected to shear deformations in the fiber direction and transverse to the fiber direction, and an equibiaxial extension. A direct comparison with stress relaxation data collected by subjecting human MCLs to shear deformation in the fiber direction is presented in order to demonstrate the predictive capabilities of the model.

Keywords Nonlinear viscoelasticity · Stress relaxation · Transversely isotropic material · Finite strain · Simple shear · Biaxial stretch · Collagenous tissue · Medial collateral ligament (MCL)

1 Introduction

Articular ligaments are soft connective tissues that connect bones to bones and stabilize the motion of joints. They are primarily composed of collagen fibers embedded in a ground substance of water, proteoglycans, and glycoproteins. Collagen fibers constitute their main load-bearing component. They are loosely organized in bundles aligned along the longitudinal axis of these ligaments (Amiel et al. 1983).

Articular ligaments are viscoelastic materials exhibiting stress relaxation, creep, and hysteresis phenomena. Stress relaxation, which is the time-dependent decrease in stress under a constant stretch, is essential to the ligaments' proper function. Indeed, this viscoelastic phenomenon acts to attenuate the stress within the stretched ligament, possibly protecting both the ligament and the surrounding structures from damage. Stress relaxation along the longitudinal axis of the ligaments has been thoroughly characterized (Provenzano et al. 2002; Hingorani et al. 2004). However, although the ligaments are primarily strained and stressed along this axis, the strain and stress distributions are three-dimensional and nonuniform due to their complex geometry and structure (Gardiner and Weiss 2003).

The importance of modeling the three-dimensional constitutive behavior of ligaments has been long recognized by the biomechanics community (Fung 1993). In recent years, several investigators have put forward constitutive models to describe the three-dimensional *elastic* response of ligaments (Lanir 1983; Hurschler et al. 1997; Gardiner and Weiss 2001; Weiss et al. 2002), but only a few have attempted to capture their three-dimensional *viscoelastic* behavior (Puso and Weiss 1998; Johnson et al. 1996; Limbert and Middleton 2004). These constitutive laws for stress relaxation have been formulated by assuming that the time-dependent and strain-dependent properties can be separated so that the normalized stress relaxation response is independent of strain. This assumption has been, however, questioned by experimental findings indicating that the normalized stress relaxation in the articular ligament's longitudinal direction is strain dependent

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(Thornton et al. 1997; Provenzano et al. 2001; Hingorani et al. 2004; Bonifasi-Lista et al. 2005).

Three-dimensional constitutive theories such as nonlinear superposition (Findley and Lai 1967), Schapery's theory (1969), and the Pipkin–Rogers integral series (1968) have been used to capture the strain-dependent normalized stress relaxation behavior of ligaments and tendons (Provenzano et al. 2002; Hingorani et al. 2004; Duenwald et al. 2009, 2010; Davis and De Vita 2012). However, these theories have been applied to simulate the constitutive behavior of these collagenous tissues solely along their fiber direction, which is the primary physiological loading direction. The first experimental studies that characterize the elastic and viscoelastic properties of articular ligaments along not only the collagen fibers' direction but also the direction transverse to these fibers (Fig. 1) have been carried out by Weiss and his research group (Quapp and Weiss 1998; Bonifasi-Lista et al. 2005). The experimental results obtained by testing these ligaments uniaxially in the transverse direction have provided crucial information about the mechanical role played by the ground substance that can be used, as a first step, to develop three-dimensional constitutive models.

In this manuscript, a three-dimensional constitutive law is presented to describe the stress relaxation behavior of articular ligaments. The constitutive model is formulated within the integral series representation proposed by Pipkin and Rogers (1968), which has been recently employed by Rajagopal and Wineman (2009) to characterize anisotropic nonlinear viscoelastic solids. The proposed model can capture the dependence of stress relaxation on strain that has been observed experimentally in collagenous tissues (Provenzano et al. 2001; Hingorani et al. 2004; Duenwald et al. 2010; Bonifasi-Lista et al. 2005) but has never been captured by three-dimensional models. Specifically, in Sect. 2, the theoretical framework set forth by Pipkin and Rogers is introduced and a constitutive relation is proposed to describe the three-dimensional stress relaxation response of articular ligaments. Published uniaxial experimental data on the elastic (Quapp and Weiss 1998) and stress relaxation behavior (Bonifasi-Lista et al. 2005) of human medial collateral ligaments (MCLs) tested along the longitudinal and transverse directions are used to determine the model parameters as described in Sect. 3. After the model parameters are identified, predictions of the stress relaxation response for simple shear in the fiber direction, simple shear transverse to the fiber direction, and equibiaxial extension are examined in Sect. 4.

2 Theoretical framework

As previously mentioned, articular ligaments are composed of an amorphous proteoglycan-rich matrix, the ground substance, which is reinforced by collagen fibers. Ligaments are assumed to be transversely isotropic since the ground substance is modeled as isotropic and the collagen fibers are preferentially aligned along the longitudinal axis of the ligament. Water is the primary component of the ground substance and accounts for more than 60 % of ligament's weight (Kannus 2000). Thus, due to the high water content, ligaments are assumed to be incompressible.

2.1 Constitutive model

The integral series representation proposed by Pipkin and Rogers (1968) was selected to describe the three-dimensional stress relaxation behavior of ligaments. Only the first term of the integral series is used so that the first Piola–Kirchhoff stress tensor, $\mathbf{P}(t)$, at any time t has the form (Rajagopal and Wineman 2009):

$$\mathbf{P}(t) = -p(t)\mathbf{F}^{-\mathrm{T}}(t) + \mathbf{F}(t)\left(\mathbf{R}[\mathbf{C}(t),0] + \int_{-\infty}^{t} \frac{\partial \mathbf{R}[\mathbf{C}(\tau), t-\tau]}{\partial(t-\tau)} d\tau\right) \tag{1}$$

where $\mathbf{F}(t)$ is the deformation gradient tensor, $\mathbf{C}(t) = \mathbf{F}^{\mathrm{T}}(t)\,\mathbf{F}(t)$ is the right Cauchy–Green deformation tensor, $\mathbf{R}[\mathbf{C}(\tau), t-\tau]$ is the tensorial relaxation function, and $p(t)$ is the Lagrange multiplier that accounts for incompressibility. Furthermore, the term $\mathbf{F}(t)\mathbf{R}[\mathbf{C}(t), 0]$ represents the instantaneous elastic contribution to the total stress at any time t. The first Piola–Kirchhoff stress tensor, $\mathbf{P}(t)$, which relates the force acting in the current configuration to the surface element in the reference configuration, is preferred in this formulation since this is the stress commonly reported in experimental studies on articular ligaments.

In Eq. (1), the use of the right Cauchy–Green deformation tensor, $\mathbf{C}(t)$, as a strain measure in the tensorial relaxation function, $\mathbf{R}[\mathbf{C}(\tau), t-\tau]$, guarantees that the principle of material frame indifference is satisfied (Truesdell et al. 2004). Assuming that the stress-free configuration is occupied at $t = 0$, in the absence of deformation, the tensorial relaxation function is identically zero. The tensorial relaxation function also needs to be a monotonically decreasing function of time to meet fading memory requirements (Truesdell et al. 2004). We explicitly note that Eq. (1) yields the general nonlinear elastic constitutive equation when time dependence is suppressed with $\mathbf{R}[\mathbf{C}(\tau)] = 2\frac{\partial W}{\partial \mathbf{C}}$ where W is the so-called strain energy density function.

Since articular ligaments are assumed to be transversely isotropic, the tensorial relaxation function $\mathbf{R}[\mathbf{C}(\tau), t-\tau]$ can be written as (Truesdell et al. 2004)

$$\mathbf{R}[\mathbf{C}(\tau), t-\tau] = k_1\mathbf{1} + k_2\mathbf{C}(\tau) + k_3\mathbf{M}\otimes\mathbf{M} + k_4\left[\mathbf{M}\otimes(\mathbf{C}(\tau)\mathbf{M}) + (\mathbf{C}(\tau)\mathbf{M})\otimes\mathbf{M}\right] \tag{2}$$

where $\mathbf{M}$ is the unit vector that defines the axis of transverse isotropy in the reference configuration and k_1, k_2, k_3, and k_4 are constitutive functions that depend on the strain invariants of $\mathbf{C}$, $I_1(\tau)$, $I_2(\tau)$, $I_4(\tau)$, $I_5(\tau)$, and $t-\tau$. The strain invariants of $\mathbf{C}$ are defined as:

$$I_1(\tau) = \text{tr}(\mathbf{C}(\tau)) , \quad I_2(\tau) = \frac{1}{2}(I_1(\tau)^2 - \text{tr}(\mathbf{C}(\tau)^2) ,$$
$$I_4(\tau) = \mathbf{M} \cdot \mathbf{C}(\tau)\mathbf{M} , \quad I_5(\tau) = \mathbf{M} \cdot \mathbf{C}(\tau)^2\mathbf{M} . \tag{3}$$

The strain invariant $I_3(\tau) = \det(\mathbf{C}(\tau)) = 1$ since the ligaments are assumed to be incompressible. Note that $I_1(\tau)$ and $I_2(\tau)$ are the strain invariants of $\mathbf{C}$ that are used to describe the isotropic response of the ground substance. The additional strain invariants, $I_4(\tau)$ and $I_5(\tau)$, account for the anisotropy generated by the presence of the collagen fiber reinforcement. By denoting the components of $\mathbf{C}$ as C_{ij} in a Cartesian coordinate system with unit vector basis $\{\mathbf{E}_1, \mathbf{E}_2, \mathbf{E}_3\}$ and assuming that $\mathbf{M} = \mathbf{E}_3$, one has that $I_4(\tau) = C_{33}$ and $I_5(\tau) = C_{13}^2 + C_{23}^2 + C_{33}^2$. It can then be observed that $I_4(\tau)$ is the square of the stretch in the fiber direction and, thus, has a clear physical interpretation. The physical interpretation of $I_5(\tau)$ cannot be given as easily; this strain invariant captures the effect of stretching and shearing in the fiber direction.

Alternatively, the tensorial relaxation function can be assumed to be given by (Rajagopal and Wineman 2009)

$$\mathbf{R}[\mathbf{C}(\tau), t-\tau] = 2\,\frac{\partial \tilde{W}(I_1(\tau), I_2(\tau), I_4(\tau), I_5(\tau), t-\tau)}{\partial \mathbf{C}} \tag{4}$$

where $\tilde{W}$ is the scalar potential density function. The constitutive functions k_1, k_2, k_3, and k_4 can be then determined from the scalar potential density function using the following relationships:

$$k_1 = 2\left(\frac{\partial \tilde{W}}{\partial I_1} + I_1(\tau)\frac{\partial \tilde{W}}{\partial I_2}\right), \quad k_2 = -2\frac{\partial \tilde{W}}{\partial I_2} ,$$
$$k_3 = 2\frac{\partial \tilde{W}}{\partial I_4} , \quad k_4 = 2\frac{\partial \tilde{W}}{\partial I_5} \tag{5}$$

where, for the ease of notation, the dependence on the strain invariants of $\mathbf{C}$ and $t-\tau$ has been dropped.

2.2 A tensorial relaxation function for articular ligaments

We adopt the following form of the tensorial relaxation function $\mathbf{R}[\mathbf{C}(\tau), t-\tau]$

$$\mathbf{R}[\mathbf{C}(\tau), t-\tau] = \mathbf{R}_{gs}[\mathbf{C}(\tau), t-\tau] + \mathbf{R}_{cf}[\mathbf{C}(\tau), t-\tau]. \tag{6}$$

where $\mathbf{R}_{gs}[\mathbf{C}(\tau), t-\tau]$ is a tensorial relaxation function representing the contribution of the *g*round *s*ubstance and $\mathbf{R}_{cf}[\mathbf{C}(\tau), t-\tau]$ is a tensorial relaxation function representing the contribution of the *c*ollagen *f*ibers. By assuming that k_1 and k_2 are functions of $I_1(\tau)$ and $t-\tau$, k_3 is a function of $I_4(\tau)$ and $t-\tau$, and k_4 is identically zero, one obtains the following relations for the tensorial relaxation functions $\mathbf{R}_{gs}$ and $\mathbf{R}_{cf}$

$$\mathbf{R}_{gs}[\mathbf{C}(\tau), t-\tau] = k_1\,(I_1(\tau), t-\tau)\,\mathbf{1} + k_2\,(I_1(\tau), t-\tau)\,\mathbf{C}(\tau) \tag{7}$$

and

$$\mathbf{R}_{cf}[\mathbf{C}(\tau), t-\tau] = k_3\,(I_4(\tau), t-\tau)\,\mathbf{M}\otimes\mathbf{M} \tag{8}$$

The above tensorial relaxation functions are fully specified once the constitutive functions k_1, k_2, and k_3 are assigned. The selection of these functions is based on the previous descriptions of the elastic and viscoelastic behavior ligaments (Fung 1993; Quapp and Weiss 1998; Weiss et al. 2002; Limbert and Middleton 2004), the details of which will be presented later (see Sect. 5). Specifically, the constitutive functions are chosen to be:

$$k_1 = \left(c_1 c_2 e^{c_2(I_1(\tau)-3)} - \frac{c_1 c_2}{2} I_1(\tau)\right) r_1(t-\tau) \tag{9}$$
$$k_2 = \frac{c_1 c_2}{2} r_1(t-\tau) \tag{10}$$
$$k_3 = \begin{cases} c_3\left(e^{c_4(I_4(\tau)-1)} - 1\right) r_2(I_4(\tau), t-\tau) & I_4(\tau) > 1 \\ 0 & I_4(\tau) \leq 1 \end{cases} \tag{11}$$

where c_1, c_2, c_3, c_4 are constants. The functions $r_1(t-\tau)$ and $r_2(I_4(\tau), t-\tau)$ are defined as:

$$r_1(t-\tau) = (1-a)\,e^{-b(t-\tau)} + a \tag{12}$$
$$r_2(I_4(\tau), t-\tau) = (1-\alpha(I_4(\tau)))\,e^{-(t-\tau)\,\beta(I_4(\tau))} + \alpha(I_4(\tau)) \tag{13}$$

where a and b are constants and $\alpha(I_4(\tau))$ and $\beta(I_4(\tau))$ are the functions of the strain invariant $I_4(\tau)$. As a result of the decomposition of the tensorial relaxation function, Eqs. (9) and (10) capture the isotropic response of the ground substance while Eq. (11) accounts for the anisotropic contribution of the collagen fibers. We explicitly note that the function k_4 from Eq. (2) is taken to be identically zero as we have suppressed dependence on $I_5(\tau)$. This assumption is common in biomechanics and reduces the number of constitutive functions which must be determined from experimental data (Holzapfel and Ogden 2009). In fact, the uniaxial data collected along the longitudinal and transverse directions of the ligaments, which are used to determine the model parameters as described in Sect. 3, are not sufficient to differentiate between the contributions of $I_4(\tau)$ and $I_5(\tau)$ (Merodio and Ogden 2005). For its simple physical interpretation, $I_4(\tau)$ is preferred over $I_5(\tau)$.

3 Identification of model parameters

Published experimental data on the elastic and stress relaxation response of human MCLs collected by subjecting specimens to uniaxial tensile tests along the longitudinal and transverse directions as schematically presented in Fig. 1 are used to determine the model parameters (Quapp and Weiss 1998; Bonifasi-Lista et al. 2005). Toward this end, the ligaments are assumed to undergo isochoric axisymmetric deformations in the longitudinal and transverse directions, and the corresponding stresses for the elastic and stress relaxation behavior are derived and presented in Sects. 3.1 and 3.2 for the longitudinal and transverse directions, respectively. These expressions are then fit to the experimental data as described in Sect. 3.3.

3.1 Axisymmetric deformation in the fiber direction

Let $\{\mathbf{E}_1, \mathbf{E}_2, \mathbf{E}_3\}$ and $\{\mathbf{e}_1, \mathbf{e}_2, \mathbf{e}_3\}$ be two sets of unit vectors that define orthonormal bases in the reference and deformed configurations, respectively. The collagen fibers are assumed to be parallel to $\mathbf{E}_3$ in the reference configuration so that $\mathbf{M} = \mathbf{E}_3$ as illustrated in Fig. 1.

The ligament is assumed to undergo an isochoric axisymmetric deformation described by

$$x_1 = \frac{1}{\lambda(t)^{1/2}} X_1\,, \quad x_2 = \frac{1}{\lambda(t)^{1/2}} X_2\,, \quad x_3 = \lambda(t)\, X_3 \tag{14}$$

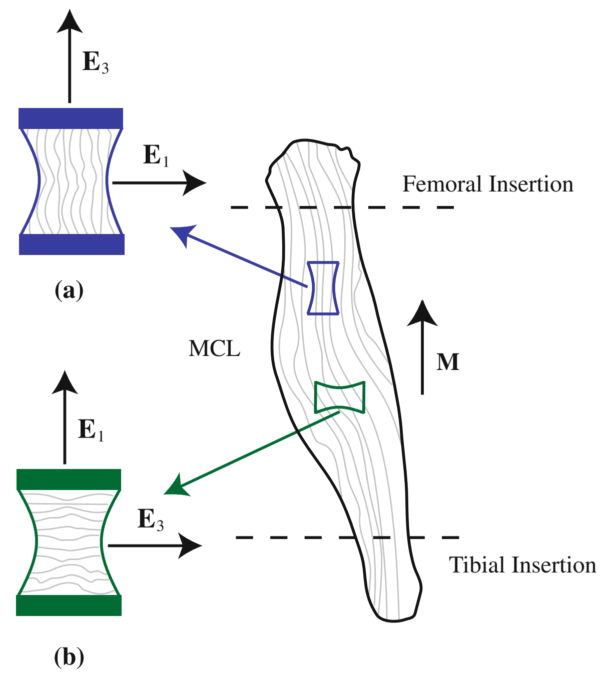

Fig. 1 Schematic of the MCL showing a sample cut in the **a** longitudinal (fiber) direction and **b** transverse direction. Note the axis of transverse isotropy in the reference configuration, **M**, coincides with $\mathbf{E}_3$

where (X_1, X_2, X_3) and (x_1, x_2, x_3) represent the Cartesian coordinates of a generic point in the reference and deformed configurations, respectively, and $\lambda(t)$ is the axial stretch that is always greater than or equal to 1. It follows that the deformation gradient tensor, $\mathbf{F}(t)$, and the right Cauchy–Green deformation tensor, $\mathbf{C}(t)$, are given by

$$\begin{aligned}\mathbf{F}(t) &= \frac{1}{\lambda(t)^{1/2}}\,\mathbf{e}_1 \otimes \mathbf{E}_1 + \frac{1}{\lambda(t)^{1/2}}\mathbf{e}_2 \otimes \mathbf{E}_2 + \lambda(t)\,\mathbf{e}_3 \otimes \mathbf{E}_3\,,\\ \mathbf{C}(t) &= \frac{1}{\lambda(t)}\,\mathbf{E}_1 \otimes \mathbf{E}_1 + \frac{1}{\lambda(t)}\,\mathbf{E}_2 \otimes \mathbf{E}_2 + \lambda(t)^2\,\mathbf{E}_3 \otimes \mathbf{E}_3\,.\end{aligned} \tag{15}$$

The first Piola–Kirchhoff stress tensor that defines the instantaneous elastic response can be computed by evaluating Eq. (1) at $t = \tau$ with the tensorial relaxation function given by Eqs. (2) and (9)–(11) with $\mathbf{F}$ and $\mathbf{C}$ given by Eq. (15). Moreover, assume that the lateral surface of the ligament is traction free so that $\mathrm{P}_{11} = \mathrm{P}_{22} = 0$. Enforcing the boundary conditions, one finds that $p = \frac{c_1 c_2}{2\lambda}\left(2e^{c_2\left(\lambda^2+2\frac{1}{\lambda}-3\right)} - \lambda^2 - \frac{1}{\lambda}\right)$. The only nonzero component of the first Piola–Kirchhoff stress tensor, P_{33}, can then be written in terms of the axial stretch λ:

$$\begin{aligned}\mathrm{P}_{33}(\lambda) &= c_3\lambda\left(e^{c_4\left(\lambda^2-1\right)} - 1\right)\\ &\quad + \frac{c_1 c_2}{2\lambda}\left(2e^{c_2\left(\lambda^2+2\frac{1}{\lambda}-3\right)} - \frac{1}{\lambda}\right)\left(\lambda^2 - \frac{1}{\lambda}\right).\end{aligned} \tag{16}$$

To model the stress relaxation response, a constant axial stretch $\lambda(t) = \hat{\lambda}$ is prescribed in the fiber direction. The stress relaxation response of the ligament can then be described using Eqs. (1), (2), (9)–(11), and (15). The lateral surface of the ligament is assumed to be traction free, and hence, $\mathrm{P}_{11}(t) = \mathrm{P}_{22}(t) = 0$. Enforcing the boundary conditions to find $p(t)$, the only nonzero component of the first Piola–Kirchhoff stress tensor, $\mathrm{P}_{33}(\hat{\lambda}, t)$, is found to be

$$\begin{aligned}\mathrm{P}_{33}(\hat{\lambda}, t) &= \frac{c_1 c_2}{2\hat{\lambda}}\left(2e^{c_2\left(\hat{\lambda}^2+\frac{2}{\hat{\lambda}}-3\right)} - \frac{1}{\hat{\lambda}}\right)\left(\hat{\lambda}^2 - \frac{1}{\hat{\lambda}}\right) r_1(t)\\ &\quad + c_3\hat{\lambda}\left(e^{c_4\left(\hat{\lambda}^2-1\right)} - 1\right) r_2(\hat{\lambda}^2, t)\end{aligned} \tag{17}$$

where the functions $r_1(t)$ and $r_2(I_4 = \hat{\lambda}^2, t)$ are defined in Eqs. (12) and (13), respectively.

3.2 Axisymmetric deformation transverse to the fiber direction

The ligament is assumed to undergo an isochoric axisymmetric deformation described by

$$x_1 = \lambda(t)\, X_1\,, \quad x_2 = \frac{1}{\lambda(t)^{1/2}} X_2\,, \quad x_3 = \frac{1}{\lambda(t)^{1/2}} X_3 \tag{18}$$

where the collagen fibers are parallel to $\mathbf{M} = \mathbf{E}_3$ in the reference configuration and $\lambda(t)$ is the axial stretch. Again, the value of the stretch is taken to be greater than or equal to 1. We note that the values of the stretches along the $\mathbf{E}_2$ and $\mathbf{E}_3$ directions are assumed to be equal since, according to selected constitutive function k_3 presented in Eq. (11), the collagen fibers do not contribute to the stress in compression. In this case, the response of the articular ligaments is governed by the ground substance, and thus, it is effectively isotropic. The deformation gradient tensor, $\mathbf{F}(t)$, and the right Cauchy–Green deformation tensor, $\mathbf{C}(t)$, are then given by

$$\mathbf{F}(t) = \lambda(t)\,\mathbf{e}_1 \otimes \mathbf{E}_1 + \frac{1}{\lambda(t)^{1/2}}\,\mathbf{e}_2 \otimes \mathbf{E}_2 + \frac{1}{\lambda(t)^{1/2}}\,\mathbf{e}_3 \otimes \mathbf{E}_3,$$
$$\mathbf{C}(t) = \lambda(t)^2\,\mathbf{E}_1 \otimes \mathbf{E}_1 + \frac{1}{\lambda(t)}\,\mathbf{E}_2 \otimes \mathbf{E}_2 + \frac{1}{\lambda(t)}\,\mathbf{E}_3 \otimes \mathbf{E}_3. \tag{19}$$

The first Piola–Kirchhoff stress tensor that defines the instantaneous elastic response for a uniaxial load in the transverse direction can be computed by evaluating Eq. (1) at $t = \tau$ with the tensorial relaxation function given by Eqs. (2) and (9)–(11) where $\mathbf{F}$ and $\mathbf{C}$ are given by Eq. (19). The traction-free boundary condition on the lateral surface requires that $\mathrm{P}_{22} = \mathrm{P}_{33} = 0$. Enforcing the boundary conditions, one finds a value for the Lagrange multiplier p. Then, the only nonzero component of the first Piola–Kirchhoff stress tensor, $\mathrm{P}_{11}(\lambda)$, is given by

$$\mathrm{P}_{11}(\lambda) = \frac{c_1 c_2}{2\lambda}\left(\lambda^2 - \frac{2}{\lambda}\right)\left(2e^{c_2\left(\lambda^2 + \frac{2}{\lambda} - 3\right)} - \frac{1}{\lambda}\right). \tag{20}$$

For a uniaxial stress relaxation in the direction transverse to the fibers, we assume that the ligament is subjected to a constant axial stretch applied transverse to the fiber direction, i.e., $\lambda(t) = \tilde{\lambda}$. The stress relaxation response of the ligament can then be described using Eqs. (1), (2), (9)–(11), and (19). The traction-free boundary condition on the lateral surface requires that $\mathrm{P}_{22}(t) = \mathrm{P}_{33}(t) = 0$. By enforcing the traction-free boundary conditions, one finds that $p(t) = \frac{c_1 c_2}{2\tilde{\lambda}}\left(2e^{c_2\left(\tilde{\lambda}^2 + 2\frac{1}{\tilde{\lambda}} - 3\right)} - \tilde{\lambda}^2 - \frac{1}{\tilde{\lambda}}\right) r_1(t)$. Then, the only nonzero component of the first Piola–Kirchhoff stress tensor, $\mathrm{P}_{11}(\tilde{\lambda}, t)$, is found to be

$$\mathrm{P}_{11}(\tilde{\lambda}, t) = \frac{c_1}{2\tilde{\lambda}}\left(\tilde{\lambda}^2 - \frac{1}{\tilde{\lambda}}\right)\left(2e^{c_2\left(\tilde{\lambda}^2 + \frac{2}{\tilde{\lambda}} - 3\right)} - \frac{1}{\tilde{\lambda}}\right) r_1(t) \tag{21}$$

where the function $r_1(t)$ is presented in Eq. (12).

3.3 Parameter identification

Due to the lack of three-dimensional experimental data, quasi-static stress–stretch data collected by performing uniaxial tensile tests on human MCLs along their longitudinal and transverse directions were used to determine the elastic material parameters (Quapp and Weiss 1998). The data were digitized from Fig. 4 of the cited manuscript by Quapp and Weiss using PlotDigitizer (v. 2.5.1). The values of the elastic material parameters, c_1, c_2, c_3, and c_4, were then determined by minimizing the sum of the squared differences between the stresses computed using Eqs. (16) and (20) and those measured experimentally simultaneously. Toward this end, the built-in minimization function in MATLAB (v. 7.12, MathWorks), *fmincon*, was employed while constraining the model parameters c_1, c_3, and c_4 to be nonnegative and $c_2 > 0.36$. These restrictions were imposed to ensure that the material response determined by the constitutive model was physically reasonable (Merodio and Ogden 2003; Bustamante and Merodio 2010; Murphy 2012). The values for the elastic material parameters were found to be $c_1 = 0.86$ MPa, $c_2 = 8.16$, $c_3 = 21.77$ MPa, and $c_4 = 3.30$. The model was able to fit well the stress–stretch data, which were collected by testing the ligamentous specimens in their longitudinal and transverse directions ($R^2 > 0.99$) (Figs. 2, 3). The nonlinear stiffening phenomenon in the stress–stretch curve of specimens oriented along the longitudinal direction (Fig. 2) and the approximately linear stress–stretch curve of specimens oriented in the transverse direction (Fig. 3) was captured.

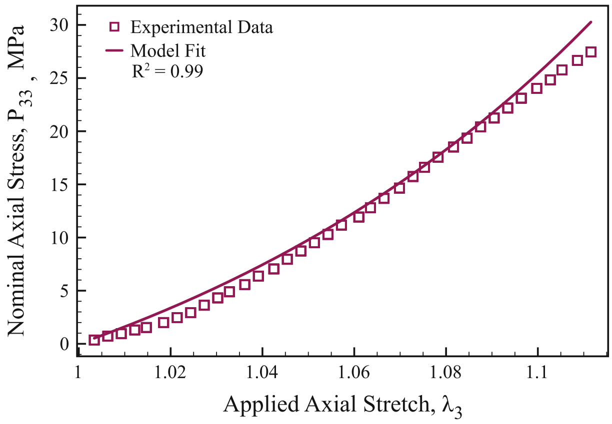

Fig. 2 Average stress–stretch experimental data for nine human MCL samples stretched along the fiber direction obtained by Quapp and Weiss (1998) and model fit with $c_1 = 0.86$ MPa, $c_2 = 8.16$, $c_3 = 21.77$ MPa, and $c_4 = 3.30$

Experiments that characterize the three-dimensional stress relaxation behavior of ligaments have yet to be performed. The most comprehensive set of experimental data on the stress relaxation response of human MCLs has been published by Bonifasi-Lista et al. (2005). Stress relaxation data were obtained by conducting uniaxial tests of ligamentous specimens in the longitudinal and transverse directions at three different strain levels. The data collected indicated that the MCL displays strain-dependent stress relaxation behavior in the longitudinal direction. However, given the lim-

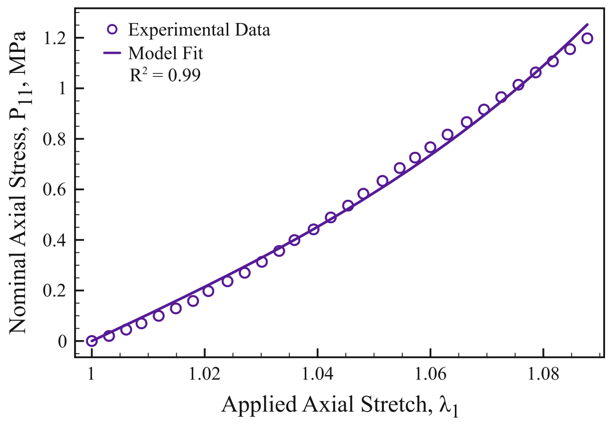

Fig. 3 Average stress–stretch experimental data for seven human MCL samples stretched along the transverse direction obtained by Quapp and Weiss (1998) and model fit with $c_1 = 0.86\,\text{MPa}$ and $c_2 = 8.16$

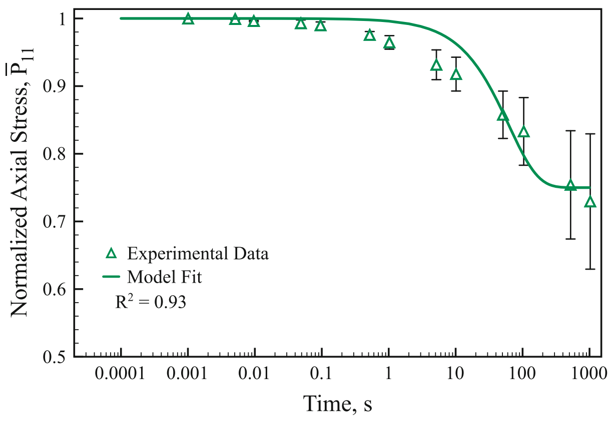

Fig. 4 Normalized experimental data (mean±SD) for five human MCLs subjected to uniaxial stress relaxation tests in the transverse direction by Bonifasi-Lista et al. (2005) and the model fit where $a = 0.75$ and $b = 0.016$ 1/s

ited number of strain levels used in the experiments, the functions $\alpha(I_4)$ and $\beta(I_4)$, which appear in Eq. (17) via Eq. (13) that capture the strain-dependent stress relaxation response, could not be computed. For this reason, these functions were set to forms previously determined by the authors for collagen fiber bundles (Davis and De Vita 2012): $\alpha(I_4) = 0.73\,e^{-14.69(I_4-1)}$ and $\beta(I_4) = 0.2084\,(I_4 - 1)$.

In order to determine the model parameters a and b, the experimental data by Bonifasi-Lista et al. (2005) obtained by subjecting specimens to uniaxial stress relaxation tests along the transverse direction at a strain level of 8 % were used. These data were digitized from Fig. 4 of the manuscript by Bonifasi-Lista et al. (2005) using PlotDigitizer. The values of the parameters a and b were found by minimizing the sum of the squared differences between the theoretical stresses in Eq. (21) and the experimentally determined stresses. The parameters were constrained to satisfy the following conditions: $0 < a < 1$ and $b > 0$. Figure 4 shows the digitized experimental data and the results of the curve fitting with $a = 0.75$ and $b = 0.016$ 1/s. Note that the predicted axial stress, $\text{P}_{11}(t)$, is normalized by its value at $t = 0$, $\text{P}_{11}(0)$, and plotted versus time. Overall, the model was able to capture the stress relaxation response in the transverse direction, but at intermediate times, when $1 < t < 10$ s, the model overpredicted the normalized stress relaxation response.

4 Model predictions

The performance of the proposed model is further evaluated by considering its predictions for simple shear in the fiber direction, simple shear transverse to the fiber direction, and equibiaxial extension. In these predictions, the material parameters and functions are fixed to those computed by fitting the uniaxial experimental data as described in the previous section. For convenience, these values are $c_1 = 0.86$ MPa, $c_2 = 8.16$, $c_3 = 21.77$ MPa, $c_4 = 3.30$, $a = 0.75$, $b = 0.016$ 1/s, $\alpha(I_4) = 0.73\,e^{-14.69(I_4-1)}$, and $\beta(I_4) = 0.2084\,(I_4 - 1)$. Note that for each of the stress relaxation predictions shown hereafter, the *normalized* stress, denoted $\overline{\text{P}}_{ij}$, is plotted to more easily visualize the influence of strain level on the stress relaxation response. The stresses are always normalized by their initial value at $t = 0$.

4.1 Simple shear in the fiber direction

Consider the isochoric deformation in the $\mathbf{E}_1$–$\mathbf{E}_3$ plane for simple shear in the fiber direction defined by

$$x_1 = X_1\,, \quad x_2 = X_2\,, \quad x_3 = \gamma(t)\,X_1 + X_3 \tag{22}$$

where the collagen fibers are parallel to $\mathbf{M} = \mathbf{E}_3$ in the reference configuration and $\gamma(t)$ is the amount of shear. The deformation gradient tensor, $\mathbf{F}(t)$, and the right Cauchy–Green deformation tensor, $\mathbf{C}(t)$, then are

$$\begin{aligned}\mathbf{F}(t) &= \mathbf{e}_1 \otimes \mathbf{E}_1 + \mathbf{e}_2 \otimes \mathbf{E}_2 + \gamma(t)\,\mathbf{e}_3 \otimes \mathbf{E}_1 + \mathbf{e}_3 \otimes \mathbf{E}_3,\\ \mathbf{C}(t) &= \left(1+\gamma(t)^2\right)\,\mathbf{E}_1 \otimes \mathbf{E}_1 + \gamma(t)\,\left(\mathbf{E}_1 \otimes \mathbf{E}_3 + \mathbf{E}_3 \otimes \mathbf{E}_1\right)\\ &\quad + \mathbf{E}_2 \otimes \mathbf{E}_2 + \mathbf{E}_3 \otimes \mathbf{E}_3\,.\end{aligned} \tag{23}$$

The collagen fibers are not stretched when the ligament is sheared in the fiber direction. Indeed, from Eq. (23) it follows that $I_4 = \text{C}_{33} = 1$. Consequently, the constitutive function k_3 defined in Eq. (11), which accounts for the contribution of the collagen fiber reinforcement, is identically zero. As a result, simple shear in the fiber direction is governed by the isotropic response of the ground substance.

The instantaneous elastic response of a ligament subjected to simple shear in the fiber direction can be computed by evaluating Eq. (1) at $t = \tau$ where the tensorial relaxation

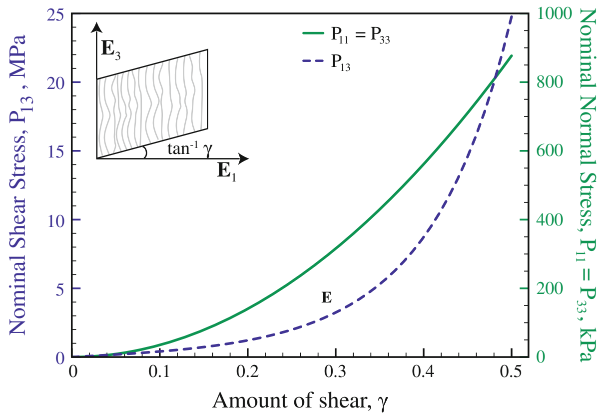

Fig. 5 Predicted shear stress, P_{13} (*left* y-axis), and normal stresses, P_{11} and P_{33} (*right* y-axis), as a function of the amount of shear, γ, for a MCL sheared in the fiber direction. The *insert* shows a schematic of the shear deformation

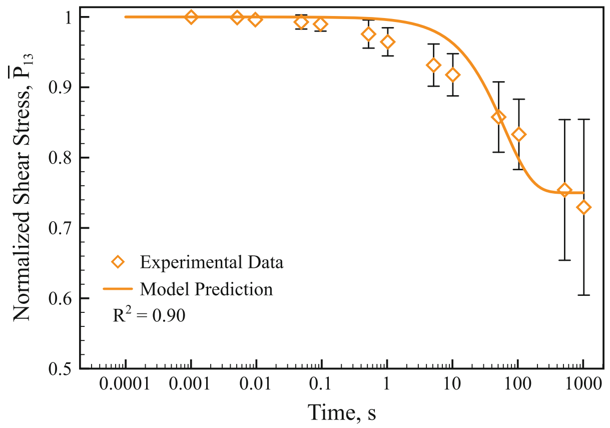

Fig. 6 Normalized stress relaxation data (mean±SD) for five human MCL samples sheared in the fiber direction and allowed to relax obtained by Bonifasi-Lista et al. (2005) for an applied shear of 0.35 and the model prediction of the normalized shear stress, $\overline{P}_{13}(t)$

function is defined by Eqs. (2) and (9)–(11). After assuming a plane stress boundary condition so that $P_{22}(t) = 0$, p can be computed and its value substituted into Eq. (1). Then, the first Piola–Kirchhoff stress tensor takes the form

$$\mathbf{P}(\gamma) = \frac{c_1 c_2 \gamma^2}{2}\,\mathbf{e}_1 \otimes \mathbf{E}_1 + \frac{c_1 c_2 \gamma}{2}\left(2e^{c_2\gamma^2} - \gamma^2 - 1\right)\mathbf{e}_1 \otimes \mathbf{E}_3 + \frac{c_1 c_2 \gamma}{2}\left(2e^{c_2\gamma^2} - 1\right)\mathbf{e}_3 \otimes \mathbf{E}_1 + \frac{c_1 c_2 \gamma^2}{2}\,\mathbf{e}_3 \otimes \mathbf{E}_3. \tag{24}$$

In Fig. 5, the shear stress, P_{13}, computed from Eq. (24) is plotted versus the amount of shear γ on the left y-axis. The resulting curve is nonlinear exhibiting the strain-stiffening phenomenon that has been observed experimentally (Weiss et al. 2002). The stiffening effect becomes more pronounced for $\gamma > 0.20$. We note that the first Piola–Kirchhoff stress tensor, $\mathbf{P}$, is not symmetric, $P_{31} \neq P_{13}$, but a similar relationship between P_{31} and γ can be obtained. The two normal stresses, P_{11} and P_{33}, required to generate a homogeneous shear are also plotted in Fig. 5 on the right y-axis. Notice that the normal stresses are an order of magnitude smaller than the shear stresses.

The first Piola–Kirchhoff stress tensor that describes the stress relaxation behavior is calculated by replacing Eqs. (2), (9)–(11), and (23) in Eq. (1) with $\gamma(t) = \hat{\gamma}$ where $\hat{\gamma}$ is a constant. By enforcing the plane stress boundary condition, which requires $P_{22}(t) = 0$, the value for the Lagrange multiplier $p(t)$ that accounts for incompressibility is determined and substituted into Eq. (1). The resulting stress tensor is:

$$\mathbf{P}(\hat{\gamma}, t) = \frac{c_1 c_2 \hat{\gamma}^2}{2} r_1(t)\,\mathbf{e}_1 \otimes \mathbf{E}_1 + \frac{c_1 c_2 \hat{\gamma}}{2}\left(2e^{c_2\hat{\gamma}^2} - \hat{\gamma}^2 - 1\right) r_1(t)\,\mathbf{e}_1 \otimes \mathbf{E}_3 + \frac{c_1 c_2 \hat{\gamma}}{2}\left(2e^{c_2\hat{\gamma}^2} - 1\right) r_1(t)\,\mathbf{e}_3 \otimes \mathbf{E}_1 + \frac{c_1 c_2 \hat{\gamma}^2}{2} r_1(t)\,\mathbf{e}_3 \otimes \mathbf{E}_3. \tag{25}$$

where the function $r_1(t)$ is presented in Eq. (12).

As noted earlier, the ligament's response to simple shear in the fiber direction is governed by the isotropic response of the ground substance. Therefore, no strain-dependent stress relaxation behavior is predicted by the proposed model for this type of deformation, as the strain-dependent relaxation behavior is attributed only to the collagen fibers. In Fig. 6, the predicted shear stress, $\overline{P}_{13}(t)$, is directly compared with experimental data published by Bonifasi-Lista et al. (2005) that were obtained by digitizing Fig. 4 in their manuscript for $\hat{\gamma} = 0.35$. Good agreement ($R^2 = 0.90$) is found between the experimental data and the model prediction. However, similar to the stress relaxation response for a stretch applied in the direction transverse to the fibers (Fig. 4), at midrange times, $t = 1 - 10$ s, the model overpredicts the value of the normalized stress.

4.2 Simple shear transverse to the fiber direction

Consider the isochoric deformation in the $\mathbf{E}_1$–$\mathbf{E}_3$ plane for simple shear transverse to the fiber direction given by

$$x_1 = X_1 + \gamma(t)\,X_3\,, \quad x_2 = X_2\,, \quad x_3 = X_3 \tag{26}$$

where $\gamma(t)$ is the amount of shear. The deformation gradient tensor, $\mathbf{F}(t)$, and the right Cauchy–Green deformation tensor, $\mathbf{C}(t)$, are

$$\mathbf{F}(t) = \mathbf{e}_1 \otimes \mathbf{E}_1 + \gamma(t)\,\mathbf{e}_1 \otimes \mathbf{E}_3 + \mathbf{e}_2 \otimes \mathbf{E}_2 + \mathbf{e}_3 \otimes \mathbf{E}_3,$$
$$\mathbf{C}(t) = \mathbf{E}_1 \otimes \mathbf{E}_1 + \gamma(t)\,(\mathbf{E}_1 \otimes \mathbf{E}_3 + \mathbf{E}_3 \otimes \mathbf{E}_1) + \mathbf{E}_2 \otimes \mathbf{E}_2 + \left(1 + \gamma(t)^2\right)\mathbf{E}_3 \otimes \mathbf{E}_3. \tag{27}$$

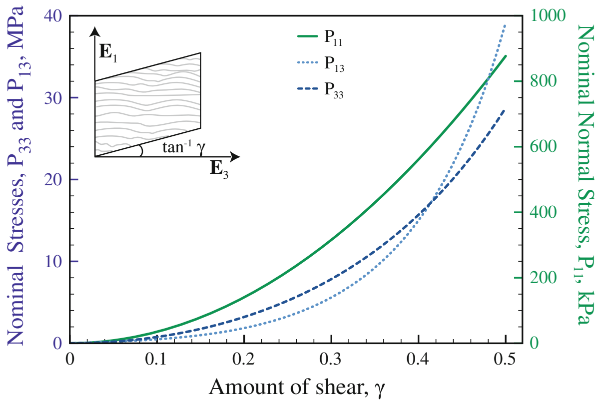

Fig. 7 Predicted shear stress, P_{13}, and normal stress, P_{33} (*left* y-axis), and the normal stress, P_{11} (*right* y-axis), as a function of the amount of shear, γ, for a MCL sheared in the direction transverse to the fibers. The *insert* shows a schematic of the shear deformation

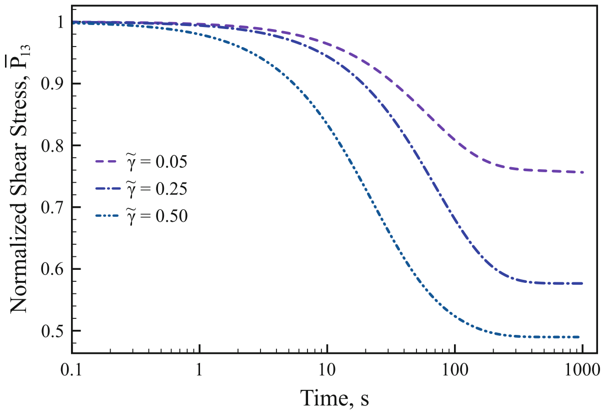

Fig. 8 Prediction of the normalized shear stress, $\overline{P}_{13}(t)$, for a MCL undergoing stress relaxation in shear transverse to the fiber direction at three levels of applied shear $\tilde{\gamma} = 0.05,\ 0.25,$ and 0.50.

The collagen fibers are stretched as the ligament is sheared. Indeed, $I_4 = C_{33} = 1 + \gamma(t)^2$. As a result, both the ground substance and the collagen fibers contribute to the ligament's response to simple shear transverse to the fiber direction.

The instantaneous elastic response to a simple shear transverse to the fiber direction is computed by evaluating Eq. (1) at $t = \tau$, with the tensorial relaxation function given by Eqs. (2) and (9)–(11) and substituting Eq. (27). The Lagrange multiplier p can be computed from the plane stress boundary condition $P_{22} = 0$. Then, the first Piola–Kirchhoff stress tensor for the instantaneous elastic response takes the form

$$\begin{aligned}\mathbf{P}(\gamma) = {} & \frac{c_1c_2}{2}\gamma^2\,\mathbf{e}_1\otimes\mathbf{E}_1 + \frac{c_1c_2\gamma}{2}\left(2e^{c_2\gamma^2} - \gamma^2 - 1\right)\mathbf{e}_3\otimes\mathbf{E}_1 \\ & + \left[c_3\gamma\left(e^{c_4\gamma^2} - 1\right) + \frac{c_1c_2\gamma}{2}\left(2e^{c_2\gamma^2} - 1\right)\right]\mathbf{e}_1\otimes\mathbf{E}_3 \\ & + \left[c_3\left(e^{c_4\gamma^2} - 1\right) + \frac{c_1c_2}{2}\gamma^2\right]\mathbf{e}_3\otimes\mathbf{E}_3. \end{aligned} \tag{28}$$

In Fig. 7, the first Piola–Kirchhoff shear stress component P_{13}, which appears in Eq. (28), is presented versus the amount of shear γ. The normal stresses, P_{11} and P_{33}, required to sustain simple shear transverse to the fiber direction are also plotted in Fig. 7. A comparison between Figs. 5 and 7 shows that the shear stress, P_{13}, and normal stress, P_{33}, are larger for the same amount of shear when the ligament undergoes simple shear transverse to the fiber direction. The increase in the shear stress, P_{13}, and normal stress, P_{33}, is due to the contribution of the collagen fibers. Notice that the normal stresses required to sustain shear transverse to the fiber direction are unequal with $P_{33} \gg P_{11}$.

Stress relaxation for simple shear transverse to the fiber direction is described by Eqs. (1), (2), and (9)–(11) with the boundary condition $P_{22} = 0$ and assuming that $\gamma(t) = \tilde{\gamma}$ is constant. The first Piola–Kirchhoff stress tensor is then

$$\begin{aligned}\mathbf{P}(\tilde{\gamma}, t) = {} & \frac{c_1c_2}{2}\tilde{\gamma}^2\, r_1(t)\,\mathbf{e}_1\otimes\mathbf{E}_1 \\ & + \frac{c_1c_2\tilde{\gamma}}{2}\left(2e^{c_2\tilde{\gamma}^2} - \tilde{\gamma}^2 - 1\right) r_1(t)\,\mathbf{e}_3\otimes\mathbf{E}_1 \\ & + \left[\frac{c_1c_2\tilde{\gamma}}{2}\left(2e^{c_2\tilde{\gamma}^2} - 1\right) r_1(t)\right. \\ & \left. + c_3\tilde{\gamma}\left(e^{c_4\tilde{\gamma}^2} - 1\right) r_2(1+\tilde{\gamma}^2, t)\right]\mathbf{e}_1\otimes\mathbf{E}_3 \\ & + \left[c_3\left(e^{c_4\tilde{\gamma}^2} - 1\right) r_2(1+\tilde{\gamma}^2, t)\right. \\ & \left. + \frac{c_1c_2}{2}\tilde{\gamma}^2\, r_1(t)\right]\mathbf{e}_3\otimes\mathbf{E}_3. \end{aligned} \tag{29}$$

where the functions $r_1(t)$ and $r_2(I_4 = 1 + \tilde{\gamma}^2, t)$ are defined in Eqs. (12) and (13).

For the deformation considered here, the collagen fibers are stretched as the ligament is sheared. Thus, since the collagen fibers are assumed to be responsible for the strain-dependent stress relaxation behavior of ligaments, the normalized stress relaxation computed via the shear stress $P_{13}(t)$ and the normal stress $P_{33}(t)$ exhibits strain dependence. In Fig. 8 the normalized shear stress, $\overline{P}_{13}(t)$, during stress relaxation is plotted for three values of the amount of shear: $\tilde{\gamma} = 0.05, 0.25, 0.50$. As the amount of shear increases, the value of the stress at equilibrium and the time to reach the equilibrium stress decrease.

4.3 Equibiaxial extension

For an isochoric equibiaxial extension along $\mathbf{E}_1$ and $\mathbf{E}_3$, the deformation is assumed to be

$$x_1 = \lambda(t)\,X_1\,,\quad x_2 = \frac{1}{\lambda^2(t)}\,X_2\,,\quad x_3 = \lambda(t)\,X_3, \tag{30}$$

where $\lambda(t)$ is the amount of stretch. The deformation gradient tensor, $\mathbf{F}(t)$, and the right Cauchy–Green deformation tensor,

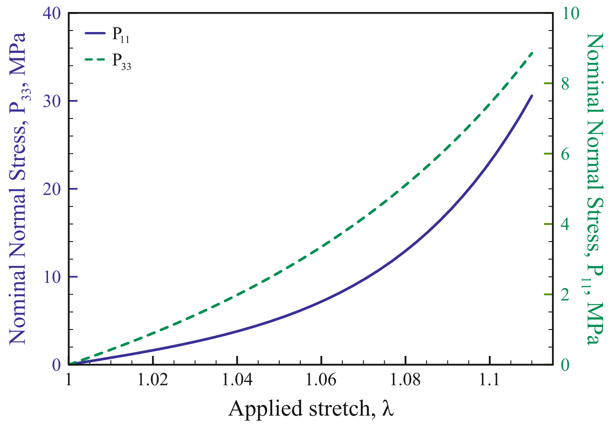

Fig. 9 Predicted stress–stretch response for the normal stress P_{33} (*left y*-axis) and the normal stress P_{11} (*right y*-axis) for a MCL subjected to an equibiaxial extension

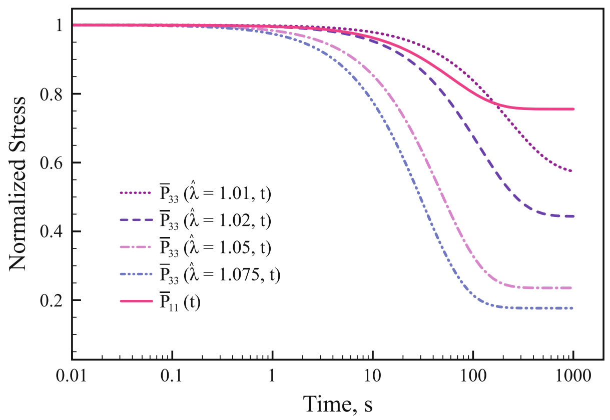

Fig. 10 Predicted normalized stress relaxation response along the $\mathbf{E}_1$ direction, $\overline{P}_{11}(t)$ (*solid line*), and the $\mathbf{E}_3$ direction, $\overline{P}_{33}(t)$ (*dashed lines*), for a MCL allowed to relax at a constant equibiaxial stretch, $\widehat{\lambda}$. In the fiber direction, the stretch level influences the normalized stress relaxation behavior so four representative stretch levels are plotted, $\widehat{\lambda} = 1.01, 1.02, 1.05$, and 1.075

$\mathbf{C}(t)$, respectively, are then given by

$$\mathbf{F}(t) = \lambda(t)\,\mathbf{e}_1 \otimes \mathbf{E}_1 + \frac{1}{\lambda(t)^2}\,\mathbf{e}_2 \otimes \mathbf{E}_2 + \lambda(t)\,\mathbf{e}_3 \otimes \mathbf{E}_3,$$
$$\mathbf{C}(t) = \lambda(t)^2\,\mathbf{E}_1 \otimes \mathbf{E}_1 + \frac{1}{\lambda(t)^4}\,\mathbf{E}_2 \otimes \mathbf{E}_2 + \lambda(t)^2\,\mathbf{E}_3 \otimes \mathbf{E}_3. \tag{31}$$

The surfaces of the ligament with outer normals $\pm\mathbf{E}_2$ are assumed to be traction free, and hence, $P_{22} = 0$. Substituting Eqs. (2), (9)–(11), and (31) into Eq. (1) at time $t = \tau$, the first Piola–Kirchhoff stress tensor for the instantaneous elastic response is computed. The Lagrange multiplier found by enforcing the traction-free boundary condition is $p = c_1 c_2 \lambda^{-4}\left(e^{c_2\left(2\lambda^2+\lambda^{-4}-3\right)} - \lambda^2\right)$. Then, the first Piola–Kirchhoff stress tensor becomes

$$\mathbf{P}(\lambda) = \frac{c_1 c_2\left(\lambda^6 - 1\right)}{2\lambda^5}\left(2e^{c_2\left(2\lambda^2+\lambda^{-4}-3\right)} - \lambda^2\right)\mathbf{e}_1 \otimes \mathbf{E}_1 + \left[\frac{c_1 c_2\left(\lambda^6 - 1\right)}{2\lambda^5}\left(2e^{c_2\left(2\lambda^2+\lambda^{-4}-3\right)} - \lambda^2\right) + c_3\lambda\left(e^{c_4\left(\lambda^2-1\right)} - 1\right)\right]\mathbf{e}_3 \otimes \mathbf{E}_3. \tag{32}$$

In Fig. 9, the normal stresses in the fiber direction, P_{33}, and in the transverse direction, P_{11}, are plotted. As expected, the normal stresses corresponding to the same stretch are much higher in the fiber direction.

The first Piola–Kirchhoff stress tensor that describes the stress relaxation response is found by using Eqs. (1), (2), (9)–(11), and (31), assuming that the stretch $\lambda(t)$ has a constant value $\widehat{\lambda}$. Again, the value for p is determined by enforcing the traction-free boundary condition on the surfaces with the outer normal $\pm\mathbf{E}_2$. The first Piola–Kirchhoff is then

$$\mathbf{P}\left(\widehat{\lambda}, t\right) = \frac{c_1 c_2\left(\widehat{\lambda}^6-1\right)}{2\widehat{\lambda}^5}\left(2e^{c_2\left(2\widehat{\lambda}^2+\widehat{\lambda}^{-4}-3\right)} - \widehat{\lambda}^2\right) r_1(t)\,\mathbf{e}_1 \otimes \mathbf{E}_1 + \left[\frac{c_1 c_2\left(\widehat{\lambda}^6 - 1\right)}{2\widehat{\lambda}^5}\left(2e^{c_2\left(2\widehat{\lambda}^2+\widehat{\lambda}^{-4}-3\right)} - \widehat{\lambda}^2\right) r_1(t) + c_3\widehat{\lambda}\left(e^{c_4\left(\widehat{\lambda}^2-1\right)} - 1\right) r_2\left(\widehat{\lambda}^2, t\right)\right]\mathbf{e}_3 \otimes \mathbf{E}_3\,. \tag{33}$$

In Fig. 10, the normalized stress relaxation response in the fiber direction, $\overline{P}_{33}(t)$, at four levels of stretch, $\widehat{\lambda} = 1.01, 1.02, 1.05$, and 1.075, is shown. The normalized stress at equilibrium and the time it takes for the normalized stress relaxation response to reach equilibrium decrease with increasing values of the applied stretch. The normalized stress relaxation response in the transverse direction, $\overline{P}_{11}(t)$, which is independent of the stretch, is also plotted in Fig. 10.

5 Discussion

A three-dimensional nonlinear constitutive relation was proposed to describe the stress relaxation response of articular ligaments. The nonlinear strain-stiffening effects, finite strains, and strain-dependent stress relaxation behavior were captured. The constitutive relation extends to three dimensions recent work done by the authors on modeling the stress relaxation response of collagen fiber bundles by accounting for the contribution of the ground substance (Davis and De Vita 2012). The model parameters were identified using published uniaxial elastic (Figs. 2, 3) and stress relaxation data (Fig. 4) obtained by testing human MCLs along the longitudinal and transverse directions (Quapp and Weiss 1998; Bonifasi-Lista et al. 2005).

The model predictions for the elastic and viscoelastic response to simple shear in the fiber direction (Figs. 5, 6), simple shear transverse to the fiber direction (Figs. 7, 8), and equibiaxial extension (Figs. 9, 10) were also presented. For simple shear in the fiber direction, the model prediction was directly compared with available empirical stress relaxation data (Bonifasi-Lista et al. 2005) (Fig. 6). The good agreement with experimental measurements suggests that the constitutive law holds promise for describing the three-dimensional viscoelastic behavior of ligaments. Further validation with three-dimensional experimental data is necessary to fully assess and, ultimately, refine the predictive capabilities of the proposed model.

The choice of the tensorial relaxation function with the constitutive functions k_1, k_2, k_3, and k_4, which describe the three-dimensional viscoelastic response of the articular ligament, was a critical component of this study. The tensorial relaxation function was defined by considering separately the contributions of the ground substance and collagen fibers (Eq. (6)). The ground substance was assumed to govern the nonlinear elastic strain-stiffening behavior observed in simple shear in the fiber direction. The collagen fibers were assumed to be responsible for the nonlinear elastic stress–stretch behavior and the strain-dependent normalized stress relaxation response of the ligaments.

The elastic response of the ground substance is obtained by setting $t = \tau$ in Eq. (7) after specifying the constitutive functions k_1 and k_2. Simple forms of these functions were considered. These include those leading to the neo-Hookean and Mooney–Rivlin models that were unable to reproduce the nonlinear strain-stiffening phenomenon observed experimentally for shear deformations in the fiber direction (Weiss et al. 2002). The constitutive functions k_1 and k_2 were then computed via Eq. (5) by adopting the strain energy proposed by Weiss et al. (2002), that is, $\tilde{W} = c_1 e^{c_2(I_1-3)} - \frac{1}{2}c_1 c_2 (I_2 - 3)$.

The elastic behavior of the collagen fibers defined by Eq. (8) for $t = \tau$ was modeled by assuming that the constitutive function k_3 is an exponential function of the strain invariant $I_4(\tau)$ when the collagen fibers are stretched, i.e., $I_4(\tau) > 1$. This form of the constitutive function can capture the strain-stiffening phenomenon exhibited by the MCL when it is axially stretched in the longitudinal direction. The function k_3 is identically zero when the collagen fibers are unstretched or compressed, i.e., $I_4(\tau) \leq 1$. This means that the collagen fibers buckle when compressed providing no contribution to the total stress. In Eq. (2), the constitutive function k_4 that depends on the strain invariant $I_5(\tau)$ was set to zero. This assumption reduces the number of material parameters that need to be determined from experimental data. For an in-depth discussion about the differences between a constitutive model for a transversely isotropic material that depends on $I_5(\tau)$ as opposed to $I_4(\tau)$, the reader is referred to the manuscript by Merodio and Ogden (2005). These authors pointed out, *inter alia*, that $I_4(\tau)$ and $I_5(\tau)$ are not independent for many plane deformations where the fiber reinforcement lies in the plane.

The normalized stress relaxation of MCLs in the transverse direction has been shown to be independent of strain (Bonifasi-Lista et al. 2005), for strain values between 8 and 12 %. For this reason, the function r_1 presented in Eq. (12), which captures the normalized stress relaxation response of the ground substance, was assumed to depend solely on time and not on strain. On the other hand, the normalized stress relaxation of collagen fibers is described by the function r_2 given in Eq. (13) that depends on time and strain. Several experimental studies on the uniaxial stress relaxation response of ligaments and tendons in the fiber direction (Hingorani et al. 2004; Provenzano et al. 2001; Bonifasi-Lista et al. 2005; Duenwald et al. 2009, 2010) including our recent study on collagen fiber bundles (Davis and De Vita 2012) support this assumption. Specifically, the functions α and β in Eq. (13) are selected to describe strain-dependent changes in the ratio of the initial stress to the equilibrium stress and the relaxation rate, respectively. It must be noted that some authors have indicated that, for very large strains, the normalized stress relaxation behavior in the fiber direction is independent of strain at least for some ligaments (Pioletti and Rakotomanana 2000; Hingorani et al. 2004). If this is indeed the case, the current model can be amended easily so that the functions α and β that appear in Eq. (13) are functions of time and strain that take constant values at these large strains.

The present study has certain limitations that need to be discussed. Due to the paucity of stress relaxation data at different strain levels for human MCLs, the functions α and β could not be determined. Therefore, they were set to be equal to those previously determined for collagen fiber bundles extracted from rat tail tendons (Davis and De Vita 2012): α was set to be a linear function and β an exponential function of I_4. Perhaps, the forms of these functions do not change significantly for human MCLs, but the values of the constants that define them are likely going to be different.

Published quasi-static stress–stretch data were used to determine the instantaneous elastic behavior of human MCLs (Quapp and Weiss 1998). In actuality, the mechanical response of ligaments is dependent on the strain rate, and thus, experimental data that capture the instantaneous elastic response of the MCLs could be quite different than the quasi-static ones used to determine the constants c_1, c_2, c_3, and c_4 (Figs. 2, 3). The strain rate dependency could be incorporated in the constitutive model by, for example, employing the approach presented by Limbert and Middleton (2004). Further experimental studies are, however, required to characterize the strain-rate-dependent behavior of articular ligaments in the fiber and transverse to the fiber directions.

Another limitation of this study, and many other studies like this on model development and validation, is the assumption that the deformations of the MCLs are homogeneous. This assumption is especially violated during shear deformations. The empirical data by Bonifasi-Lista et al. (2005) used to validate the proposed model were collected using an apparatus that could not provide normal tractions on all of the inclined faces of the specimens. These normal tractions are required to obtain a homogeneous shear deformation (Truesdell et al. 2004; Horgan and Murphy 2011). Only for small strains can a relatively homogeneous shear deformation of a nonlinear elastic specimen be produced without supplying the required normal stresses. For this reason, the spatial variation in the strain field of articular ligaments needs to be measured using optical techniques, such as the digital image correlation methods (Zhang and Arola 2004), and compared with a finite element implementation of the constitutive model for complete model validation.

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References

Amiel D, Frank C, Harwood F, Fronek J, Akeson W (1983) Tendons and ligaments: a morphological and biochemical comparison. J Orthop Res 1(3):257–265

Bonifasi-Lista C, Lakez SP, Small MS, Weiss JA (2005) Viscoelastic properties of the human medial collateral ligament under longitudinal, transverse and shear loading. J Orthop Res 23(1):67–76

Bustamante R, Merodio J (2010) On simple constitutive restrictions for transversely isotropic nonlinearly elastic materials and isotropic magneto-sensitive elastomers. J Eng Math 68(1):15–26

Davis FM, De Vita R (2012) A nonlinear constitutive model for stress relaxation in ligaments and tendons. Ann Biomed Eng 40(12):2541–2550

Duenwald SE, Vanderby R, Lakes RS (2009) Viscoelastic relaxation and recovery of tendon. Ann Biomed Eng 37(6):1131–1140

Duenwald SE, Vanderby R, Lakes RS (2010) Stress relaxation and recovery in tendon and ligament: experiment and modeling. Biorheology 47(1):1–14

Findley WN, Lai JSY (1967) A modified superposition principle applied to the creep of nonlinear viscoelastic material under abrupt changes in state of combined stress. Trans Soc Rheol 11(3):361–380

Fung YC (1993) Biomechanics, mechanical properties of living tissues, 2nd edn. Springer, New York

Gardiner JC, Weiss JA (2001) Simple shear testing of parallel-fibered planar soft tissues. J Biomech Eng Trans ASME 123(2):170–175

Gardiner JC, Weiss JA (2003) Subject-specific finite element analysis of the human medial collateral ligament during valgus knee loading. J Orthop Res 21(6):1098–1106

Hingorani RV, Provenzano PP, Lakes RS, Escarcega A, Vanderby R (2004) Nonlinear viscoelasticity in rabbit medial collateral ligament. Ann Biomed Eng 32(2):306–312

Holzapfel GA, Ogden RW (2009) On planar biaxial tests for anisotropic nonlinearly elastic solids: a continuum mechanical framework. Math Mech Solids 14(5):474–489

Horgan CO, Murphy JG (2011) Simple shearing of soft biological tissues. Proc R Soc A-Math Phys Eng Sci 467:760–777

Hurschler C, Loitz-Ramage B, Vanderby R (1997) A structurally based stress-stretch relationship for tendon and ligament. J Biomech Eng Trans ASME 119(4):392–399

Johnson GA, Livesay GA, Woo SLY, Rajagopal KR (1996) A single integral finite strain viscoelastic model of ligaments and tendons. J Biomech Eng Trans ASME 118(2):221–226

Kannus P (2000) Structure of the tendon connective tissue. Scand J Med Sci Sports 10(6):312–320

Lanir Y (1983) Constitutive equations for fibrous connective tissues. J Biomech 16(1):1–12

Limbert G, Middleton J (2004) A transversely isotropic viscohyperelastic material—application to the modeling of biological soft connective tissues. Int J Solids Struct 41(15):4237–4260

Merodio J, Ogden RW (2003) A note on strong ellipticity for transversely isotropic linearly elastic solids. Q J Mech Appl Math 56(4):589–591

Merodio J, Ogden RW (2005) Mechanical response of fiber-reinforced incompressible non-linearly elastic solids. Int J Non-Linear Mech 40:213–227

Murphy JG (2012) Tension in the fibres of anisotropic non-linearly hyperelastic materials. some stability results and constitutive restrictions. Int J Solids Struct 50(2):423–428

Pioletti DP, Rakotomanana LR (2000) On the independence of time and strain effects in the stress relaxation of ligaments and tendons. J Biomech 33(12):1729–1732

Pipkin AC, Rogers TG (1968) A non-linear integral representation for viscoelastic behaviour. J Mech Phys Solids 16(1):59–72

Provenzano PP, Lakes RS, Keenan T, Vanderby R (2001) Nonlinear ligament viscoelasticity. Ann Biomed Eng 29(10):908–914

Provenzano PP, Lakes RS, Corr DT, Vanderby R (2002) Application of nonlinear viscoelastic models to describe ligament behavior. Biomech Model Mechanobiol 1(1):45–57

Puso MA, Weiss JA (1998) Finite element implementation of anisotropic quasi-linear viscoelasticity using a discrete spectrum approximation. J Biomech Eng Trans ASME 120(1):62–70

Quapp KM, Weiss JA (1998) Material characterization of human medial collateral ligament. J Biomech Eng Trans ASME 120:757–763

Rajagopal KR, Wineman AS (2009) Response of anisotropic nonlinearly viscoelastic solids. Math Mech Solids 14(5):490–501

Schapery R (1969) On the characterization of nonlinear viscoelastic materials. Polym Eng Sci 9(4):295–310

Thornton GM, Oliynyk A, Frank CB, Shrive NG (1997) Ligament creep cannot be predicted from stress relaxation at low stress: a biomechanical study of the rabbit medial collateral ligament. J Orthop Res 15(5):652–656

Truesdell C, Noll W, Antman SS (2004) The non-linear field theories of mechanics, vol 3. Springer, Berlin

Weiss JA, Gardiner JC, Bonifasi-Lista C (2002) Ligament material behavior is nonlinear, viscoelastic and rate-independent under shear loading. J Biomech 35(7):943–950

Zhang D, Arola DD (2004) Applications of digital image correlation to biological tissues. J Biomed Opt 9(4):691–699