Original paper

Biomechan Model Mechanobiol 1 (2002) 45 – 57 © Springer-Verlag 2002
DOI 10.1007/s10237-002-0004-1

Application of nonlinear viscoelastic models to describe ligament behavior

P. P. Provenzano, R. S. Lakes, D. T. Corr, R. Vanderby Jr.

Abstract Recent experiments in rat medial collateral ligament revealed that the rate of stress relaxation is strain dependent and the rate of creep is stress dependent. This nonlinear behavior requires a more general description than the separable quasilinear viscoelasticity theory commonly used in tissue biomechanics. The purpose of this study was to determine whether the nonlinear theory of Schapery or the modified superposition method could adequately model the strain-dependent stress-relaxation behavior of ligaments. It is shown herein that both theories describe available nonlinear experimental ligament data well and hence can account for both elastic and viscous nonlinearities. However, modified superposition allows for a more direct interpretation of the relationship between model parameters and physical behavior, such as elastic and viscous nonlinearities, than does Schapery's theory. Hence, the modified superposition model is suggested to describe ligament data demonstrating both elastic nonlinearity and strain-dependent relaxation rate behavior. The behavior of the modified superposition model under a sinusoidal strain history is also examined. The model predicts that both elastic and viscous behaviors are dependent on strain amplitude and frequency.

1 Introduction

Ligaments display time-dependent and history-dependent mechanical behavior characteristic of viscoelastic materials. Viscoelastic behavior has been observed and studied in cells (Bausch et al. 1999; Guilak 2000; Guilak et al. 1999, 2000; Heidemann et al. 1999; Trickey et al. 2000) and a number of biologic tissues such as articular cartilage (Mak 1986; Woo et al. 1980), bone (Lakes and Katz 1979; Lakes et al. 1979), skeletal muscle (Best et al. 1994), cardiovascular tissue (Rousseau et al. 1983; Sauren et al. 1983), tendon (Atkinson et al. 1999; Graf et al. 1994), and ligament (Haut and Little 1969; Provenzano et al. 2001; Thornton et al. 1997; Woo 1982, Woo et al. 1981). Structural, phenomenological, and continuum models have been formulated to describe these viscoelastic behaviors (Bingham and DeHoff 1979; Corr et al. 2001; Decraemer

Received: 6 November 2001 / Accepted: 11 January 2002

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This work was funded by the N.S.F. (grant CMS-9907977) and NASA (grant NAG9-1152).

et al. 1980; Dehoff 1978; Egan 1987; Fung 1972; Johnson et al. 1996; Lakes and Vanderby 1999; Lanir 1979, 1980, 1983; Sanjeevi et al. 1982; Shoemaker et al. 1986; Viidik 1968). The most commonly applied model of viscoelastic behavior in biomechanics has been the quasilinear viscoelasticity (QLV) model of Fung (1972). This model has been particularly useful in describing experimental behavior in soft tissues (Best et al. 1994; Carew et al. 1999, 2000; Fung 1972; Sauren and Rousseau 1983; Sauren et al. 1983; Thornton et al. 1997; Woo 1982; Woo et al. 1980, 1981) and has been shown to describe ligament relaxation behavior at a single fixed strain level very well (Woo 1982; Woo et al. 1981).

A recent study (Provenzano et al. 2001) in rat medial collateral ligament revealed that within the strain-stiffening "toe" region and early portions of the linear region of the stress–strain curve, stress relaxation, and creep behavior are nonlinear functions of strain and stress, respectively. The rate of stress relaxation decreases with increasing strain and the rate of creep decreases with increasing stress. Similar strain-dependent relaxation rate behavior has been reported in the fibrocartilage zone of rabbit tendon tested in compression (Haridas et al. 2001). The behavior in these data sets cannot be robustly described using QLV, since in the separable formulation the time-dependent behavior is independent of stress or strain. Hence, the same rate of relaxation or creep would be predicted regardless of strain or stress level (Provenzano et al. 2001). Although each curve in the data set could be individually fit with separate moduli and a range obtained (as was demonstrated by Haridas et al. 2001), with QLV a single modulus cannot describe the stress- or strain-dependent rate behavior. A more general formulation is therefore required for these data.

Thornton et al. (1997) reported that stress relaxation proceeds more rapidly than creep and demonstrated that neither a linear nor a QLV theory was able to phenomenologically model both behaviors with interrelated constitutive coefficients. Such behaviors can be described, however, using the single integral form of nonlinear superposition with interrelated coefficients for relaxation and creep as shown by Lakes and Vanderby (1999), or by incorporating collagen fiber recruitment when predicting creep from stress relaxation as shown by Thornton et al. (2001). These studies examined the relaxation–creep interrelation at only one level of coupled strain–stress, therefore it is not yet known if these models can account for the strain- or stress-dependent behavior described above, or if a more general formulation is required.

Many reasonably general constitutive models such those by Schapery (1969), Lai and Findley (1968), Christensen (1980), Pipkin and Rogers (1968) and the modified superposition (also commonly referred to as nonlinear superposition) method (Findley et al. 1976; Lai and Findley 1968; Lakes 1998) have been proposed to describe nonlinearly viscoelastic materials. The Schapery single integral approach has been shown to be accurate and adaptable (Dillard et al. 1987; Lou and Schapery 1971; Touti and Cederbaum 1997) and modified superposition is general and also allows the relaxation function to depend on strain. These models have not been used to describe ligaments, but some have been used for polymers, and their formulations show potential for ligament mechanics. Therefore, the objective of this study is to determine whether the theory of Schapery (1969) or the modified superposition method (Findley et al. 1976) can adequately model the stress- and strain-dependent creep and relaxation behavior of ligaments.

2 Analysis

Because this study investigates the application of existing viscoelastic models to existing experimental ligament data, all models are one-dimensional (1-D) and employ small (infinitesimal) strains and engineering stress. These theories are generalizable to 3-D but this is not currently necessary or desirable since we are dealing with a simple tension, 1-D experiment. As such, it is not meaningful to distinguish isotropy from anisotropy or compressible from incompressible. Regarding the chosen stress and strain formulations, the maximum strain in the ligament data is about 0.05; hence, the error associated with using small strain can be neglected.

Linear and quasilinear viscoelastic theory

Linear viscoelastic behavior is commonly described using the Boltzmann superposition integral:

$$\sigma(t) = \int_0^t E(t-\tau)\frac{\mathrm{d}\varepsilon(\tau)}{\mathrm{d}\tau}\mathrm{d}\tau \ , \tag{1}$$

in which $\sigma(t)$ is the stress, $E(t)$ is the time-dependent relaxation function, $\varepsilon(t)$ is the strain, and τ is the variable of integration. A complementary relation can be obtained for creep behavior. This formulation does not allow the relaxation or compliance function to depend on strain or stress, nor does it allow for elastic nonlinearity. Therefore, a more general formulation is required for nonlinear materials such as ligament.

QLV (Fung 1972) accounts for elastic nonlinearity of the stress–strain behavior:

$$\sigma(\varepsilon, t) = \int_0^t E_t(t-\tau) \frac{d\sigma}{d\varepsilon} \frac{d\varepsilon(\tau)}{d\tau} d\tau \quad (2)$$

whereby, the relaxation function is separable into a function of time and a function of strain, i.e., $E(t,\varepsilon) = E_t(t)g(\varepsilon)$. The function $g(\varepsilon)$ accounts for strain-dependent elastic nonlinearity ($d\sigma/d\varepsilon$ in Eq. 2). With QLV, the time-dependent portion of behavior $E_t(t)$ is independent of strain. Considering the strain history to be controlled by a Heaviside step function (i.e., $\varepsilon(t) = \varepsilon_0 H(t)$), the derivative of strain in Eq. (2) will become a delta function and Eq. (2) becomes $\sigma(\varepsilon_0,t) = \varepsilon_0 E(t)g(\varepsilon_0)$. Thus, stress is strain dependent but the time-dependent portion of the modulus, and thus, time-dependent stress behavior, is independent of strain level. Elastic nonlinearity and time dependence can be discriminated graphically. When examining stress-relaxation data, the strain-dependent elastic nonlinearity is revealed by plotting isochronal stress versus strain, while the time dependence is related to the shape and strain dependence of the relaxation curves. This graphical representation can be studied on a log–log plot of stress versus time where non-strain-dependent curves will have the same shape, or if the modulus is in the form of a power law, the same slope (Provenzano et al. 2001). A similar formulation can be obtained for creep behavior. Although QLV accounts for elastic nonlinearity, the time dependence is independent of strain. The QLV theory would therefore predict the same relaxation rate regardless of applied strain level. Hence, a more general formulation is required to account for the strain-dependent relaxation rate behavior seen in ligament.

The QLV model has been used for many years; limitations in this model have not been apparent in part because many experiments were designed under the assumption of QLV. Such experiments in principle cannot robustly test the QLV model. A common experimental modality is to conduct a single test at constant strain rate to determine the nonlinearity, and a single creep or relaxation test to determine the time dependence. Such an experiment cannot distinguish between a separable kernel and a nonseparable kernel within the framework of single integral models.

2
Schapery's single integral nonlinear theory

Schapery's nonlinear viscoelastic theory can be derived using principles of irreversible thermodynamics (Lou and Schapery 1971; Schapery 1966, 1969). When strain is treated as the independent state variable and the case of uniaxial loading is considered, Schapery's theory reduces to a single integral expression:

$$\sigma(\varepsilon, t) = h_e(\varepsilon) E_e \varepsilon + h_1(\varepsilon) \int_0^t \Delta E(\rho(t) - \rho'(\tau)) \frac{dh_2(\varepsilon)\varepsilon}{d\tau} d\tau \quad (3)$$

with the reduced time, ρ, defined as,

$$\rho = \int_0^t \frac{dt'}{a_e[\varepsilon(t')]} \quad (4)$$

and reduced time variable of integration ρ',

$$\rho' = \int_0^\tau \frac{dt'}{a_e[\varepsilon(t')]} \quad (5)$$

In Eq. (3), E_e is the equilibrium, or final, value of the elastic modulus, and ΔE is the transient modulus. Since "final" refers to behavior at infinite time, which is not available to a mortal observer, the quantity E_e for practical purposes is the modulus value of the last data point in the experimental time frame. Ideally, the final data point would occur after some "steady-state" behavior has been approached. The terms h_e, h_1, h_2, and a_e are strain-dependent material properties (a_e is a function of strain and time and may also be temperature dependent) that have thermodynamic significance; variations in the first three terms are due to third- and higher-order strain effects in the Helmholtz free energy and changes in a_e are related to strain and temperature influence on entropy and free energy (Schapery 1966, 1969). Schapery (1969) presented a complementary relation for creep in which nonlinear stress-dependent terms are related to Gibb's free energy.

The embedded function a_e in the reduced time expressions (Eqs. 4 and 5) is used to shift the viscoelastic time scale. The variable a_e relates to the effect of temperature. In particular, for some materials, notably polymers, a change in temperature gives rise to an acceleration or retardation of time-dependent processes. This is referred to time–temperature superposition. Materials that behave in this way are referred to as thermorheologically simple. Composites, whether they are synthetic or biological (such as bone), tend not to obey time–temperature superposition (Lakes 1998). Temperature dependence is not considered in this article for ligament.

When $h_e = h_1 = h_2 = a_e = 1$, Eq. 3 reduces to the Boltzmann superposition principle (Eq. 1). However, when implementing the above theory to nonlinear behavior, one or more of the strain-dependent functions (h_e, h_1, h_2, a_e), but not all, can be assumed to equal unity (Dillard et al. 1987; Lou and Schapery 1971; Schapery 1969; Touti and Cederbaum 1997). In polymers and fibrous composite materials in isothermal settings it is common to set h_1 and a_e to unity (Dillard et al. 1987; Lou and Schapery 1971; Schapery 1969; Touti and Cederbaum 1997); this case will be applied in this study.

In previous applications of Schapery's theory with constant strain (after initial ramp time), the transient modulus has been modeled using a power law formulation (Dillard et al. 1987; Lou and Schapery 1971; Schapery 1969), which has been shown to describe ligament viscoelastic behavior well (Lakes and Vanderby 1999; Provenzano et al. 2001). Therefore, in the constant strain application of Schapery's theory the form of the transient modulus will be modeled as a power law:

$$\Delta E(\rho) = \mathrm{C}\rho^{\mathrm{n}} \quad , \tag{6}$$

where C and n are assumed to be material constants at any strain level, for a constant temperature (Dillard et al. 1987). When Eq. (6) is substituted into Eq. (3) the stress is:

$$\sigma(\varepsilon, t) = h_e(\varepsilon)E_e\varepsilon + h_1(\varepsilon)\mathrm{C}\int_0^t (\rho - \rho')^{\mathrm{n}} \frac{\mathrm{d}h_2(\varepsilon)\varepsilon}{\mathrm{d}\tau}\mathrm{d}\tau \quad . \tag{7}$$

Substituting a Heaviside function into Eq. (7) for a particular strain, ε_0, and setting $h_1 = a_e = 1$, as discussed above, results in:

$$\sigma(\varepsilon_0, t) = h_e E_e \varepsilon_0 + h_2 \mathrm{C} \varepsilon_0 t^{\mathrm{n}} \quad . \tag{8}$$

Noting that $E_e\varepsilon$ and $\mathrm{C}\varepsilon t^{\mathrm{n}}$ of Eq. (8) take the role of final and transient stresses, respectively, that E_e and ε are known from experimental data, and that C and n are constants determined by curve fitting, it can be seen that Eq. (8) predicts the strain-dependent elastic and rate behavior with two parameters that are functions of strain, h_e and h_2.

Modied superposition method

The single integral formulation of the modified superposition method (Findley et al. 1976; Lakes 1998) allows the relaxation function to depend on strain level:

$$\sigma(\varepsilon, t) = \int_0^t E(t - \tau, \varepsilon(\tau)) \frac{\mathrm{d}\varepsilon(\tau)}{\mathrm{d}\tau}\mathrm{d}\tau \quad . \tag{9}$$

A similar stress-dependent compliance formulation exists for creep. The form of the relaxation function will be chosen as a nonseparable strain-dependent power law:

$$E(\varepsilon, t) = A(\varepsilon)t^{B(\varepsilon)} \quad . \tag{10}$$

The function $A(\varepsilon)$ represents the initial modulus (E_0), which can be obtained from a stress–strain curve or isochronal curve describing the nonlinear elastic behavior. The function $B(\varepsilon)$ describes the strain-dependent rate of stress relaxation and can take the form $B(\varepsilon)=g(\varepsilon)n_0$, where n_0 is some initial relaxation rate and $g(\varepsilon)$ accounts for strain-dependent nonlinearity in relaxation rate. Substituting a Heaviside function, as described above, into Eq. (9) results in:

$$\sigma(\varepsilon, t) = E_0 \varepsilon t^{g(\varepsilon)n_0} = \sigma_0 t^{g(\varepsilon)n_0} \tag{11}$$

where E_0 and σ_0 represent isochronal values of the tangent modulus and stress, respectively, and can be functions of strain to account for nonlinearities in the elastic response. In addition, Eq. (11) can take on a more predictive form once relaxation or creep rates over a range of strain or stress values are obtained, i.e., the dependence of the rate B as a function of strain or stress is known. Stress–strain or isochronal curves can be used to obtain the initial modulus or stress terms, $A(\varepsilon)$, and a polynomial can be fit to the rate range to obtain the function $B(\varepsilon)$; future mechanical behavior within the strain range can then be predicted. Hence, the nonseparable form of modified superposition is able to represent both the elastic and strain-dependent rate nonlinearities that are experimentally observed.

In addition to the Heaviside function, a more complex strain-history function will be examined. A sinusoidal strain history of the form:

$$\varepsilon(t) = \frac{\varepsilon_0}{2}[1 - \cos(\alpha\pi t)] \tag{12}$$

was chosen, where ε_0 is the strain amplitude and α is related to the frequency (α is twice the frequency; Fig. 1). The resulting form of Eq. (9) is:

$$\sigma(\varepsilon, t) = \frac{\pi\alpha\varepsilon_0}{2} \int_0^t A(\varepsilon)(t - \tau)^{B(\varepsilon)} \sin(\alpha\pi\tau)\mathrm{d}\tau \quad . \tag{13}$$

Experimental data

Each of the preceding nonlinear models will be applied to previously published experimental stress-relaxation data from rat medial collateral ligament (Provenzano et al. 2001). The experimental methods for these data are described in detail elsewhere, and therefore will only be

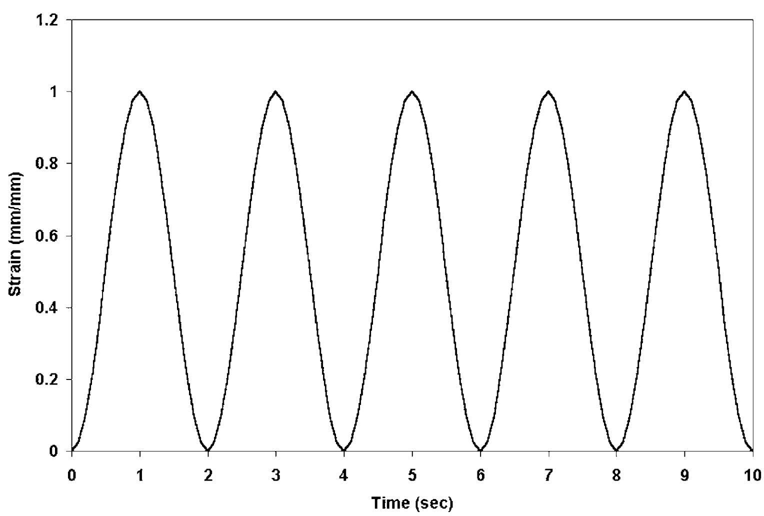

Fig. 1. Behavior of the sinusoidal strain history function applied within the framework of modified superposition. The strain amplitude, ε_0, has been chosen as unity in this example

discussed briefly. A total of six ligaments from separate animals composed the experimental stress-relaxation group. Multiple uniaxial stress-relaxation tests were performed on each ligament. Each tissue was tested at constant strain (ramp time = 0.32 s) for 100 s at levels of strain below the damage threshold of ~5% for this method of testing (Provenzano et al. 2002), allowed to recover for at least 10 times the length of the test while remaining hydrated, then tested again at a different strain level. The test order was randomized, and the area of the tissue was calculated by optical measurement of the tissue width and thickness, and assuming an elliptical cross-section. Strain was measured on the ligament. Acquired force, strain, and time were synchronized. Engineering stress and strain were then calculated and stress plotted versus time. The first data point was plotted at 10 times the ramp time in order to reduce transient effects due to tissue loading. Therefore, the initial stress amplitudes represent isochronal data. A power law, t^n, was fit to the experimental data using KaleidaGraph (Synergy Software, Inc., Reading, Penn., USA) graphical software. Results showed the rate of relaxation to be dependent upon the level of strain. Both Schapery's theory and the modified superposition method will be applied to model typical data displaying this nonlinear behavior. Model parameters were determined by programming each of the models into KaleidaGraph and applying them to the experimental data described above.

To the authors' knowledge no tests exist revealing strain-dependent relaxation behavior during cyclic testing. Hence, the stress resulting from a sinusoidal strain history in Eq. (13) demonstrates model behavior only. Eq. (13) was programmed into MATLAB (The MathWorks, Inc., Natick, Mass., USA) and solved for strain-amplitude values less than 5%. These strain values were chosen because, as discussed above, this region is known to display nonlinear strain-dependent relaxation rate behavior under constant strain.

3 Results

Application of Schapery's theory to ligament data

The nonlinear theory of Schapery (1969) (Eqs. 3–8) was applied to experimental stress-relaxation data from rat MCLs (Fig. 2). The data demonstrate that the rate of stress relaxation is significantly dependent upon strain, with decreasing relaxation rate with increasing tissue strain. Schapery's nonlinear theory was able to fit the data well at all strain levels: $\varepsilon = 0.82\%$, $R^2 = 0.83$; $\varepsilon = 1.74\%$, $R^2 = 0.96$; $\varepsilon = 2.38\%$, $R^2 = 0.99$; $\varepsilon = 3.74\%$, $R^2 = 0.98$. Hence, Schapery's theory accounts for the elastic nonlinearity and strain-dependent relaxation rate nonlinearity observed in ligament. The initial isochronal stress amplitudes provided by Schapery's theory are 1.26, 5.48, 9.91, and 14.88 MPa for 0.82, 1.74, 2.38, and 3.74% strains, respectively, and as such reveal the elastic nonlinearity in ligament. Decreases in the strain-dependent $h_2(\varepsilon)$ term with increasing strain can be related to decreasing relaxation rate with increasing strain. The constants C and n, determined by fitting the first ($\varepsilon = 0.82\%$) curve with h_e and h_2 equal to unity, are 118.85 and –0.5, respectively. The strain-dependent terms for the remaining three stress-relaxation curves at 1.74, 2.38,

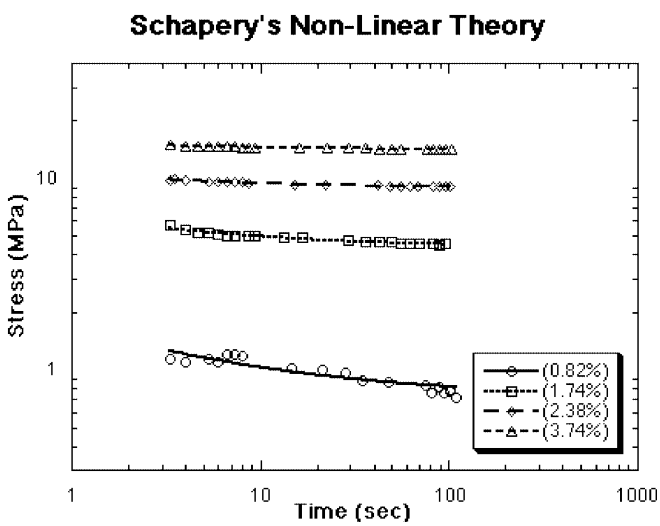

Fig. 2. Application of Schapery's nonlinear viscoelasticity theory (*curves*) to experimental stress-relaxation results of Provenzano et al. (2001) (*points*) for multiple testing at various strain levels of a single rat ligament. R^2 = 0.83, 0.96, 0.99, and 0.98 for ε = 0.82%, 1.74%, 2.38%, and 3.74%, respectively

and 3.74% strain are respectively, h_e = 0.946, 0.977, and 0.990; h_2 = 1.028, 0.678, and 0.331. Hence, $h_2(\varepsilon)$ decreased by approximately 68% from 1.74 to 3.74% strain indicating a reduction in relaxation rate with increasing strain. In addition to stress levels and rate information, a range of moduli information can be obtained from Schapery's theory by employing an alternate form of Eq. (3):

$$\sigma(\varepsilon, t) = h_0(\varepsilon)E_0\varepsilon - h_3(\varepsilon)\int_0^t E_R(\rho(t) - \rho'(\tau))\frac{\mathrm{d}h_4(\varepsilon)\varepsilon}{\mathrm{d}\tau}\mathrm{d}\tau \tag{14}$$

where E_0 is the initial time-independent elastic modulus, E_R is the relaxation modulus, and the functions h_0, h_3, and h_4 are the strain-dependent terms. Examining Eqs. (3) and (14) it is seen that the moduli are related as follows (Findley et al. 1976):

$$E_0 = E_R + \Delta E + E_e \tag{15}$$

Applying this relation to the data in Fig. 2, the elastic moduli at 10 s are found to be 138, 299, 400, and 401 MPa for 0.82, 1.74, 2.38, and 3.74% strain, respectively. These data (elastic modulus at a time point within the data set) can be obtained by calculating the relaxation modulus and subtracting it from the initial elastic modulus, or by calculating the transient modulus and adding it to the final elastic modulus. Finally, examination of the model data demonstrates that although the transient modulus is in the form of a power-law, the final plot is not linear on a log–log scale due to the additive nature of Schapery's formulation. The exponent in the power law is a constant and the nonlinearity is accounted for by two nonlinear terms, h_e and h_2, each multiplied by a set of constants and added as seen in Eq. (8). Hence, nonlinearity in the rate of relaxation with strain is not determined by changes in the exponent of the power law but by two additive terms where the magnitude of h_2 affects the weight of the power law term and thus strongly influences the predicted rate of relaxation.

Application of the modied superposition method to ligament data

Modified superposition theory (Eqs. 9–11) was applied to experimental stress-relaxation data from rat MCLs (Fig. 3). As stated above, these data demonstrate that the rate of stress relaxation decreases significantly (more than an order of magnitude) with increasing tissue strain. Modified superposition theory fits the data well for all strain levels: ε = 0.82%, R^2 = 0.91; ε = 1.74%, R^2 = 0.91; ε = 2.38%, R^2 = 0.96; ε = 3.74%, R^2 = 0.95. Application of the model resulted in σ_0 values of 1.45, 5.72, 10.10, and 15.25 MPa for 0.82, 1.74, 2.38, and 3.74% strain, respectively, revealing elastic nonlinearity. The rate term, $B(\varepsilon)$, can be seen to decrease in magnitude as strain increases (–0.14, –0.054, –0.026, and –0.012 for 0.82, 1.74, 2.38, and 3.74% strain, respectively) indicating nonlinearity in the rate of relaxation with strain. This nonlinearity can easily be visualized as straight-line segments on a log–log plot (Fig. 3). Having obtained the above values, the isochronal stress amplitudes at the first isochronal data points can be calculated as 1.23, 5.36, 9.80, and 14.80 MPa for 0.82, 1.74, 2.38, and 3.74% strain, respectively.

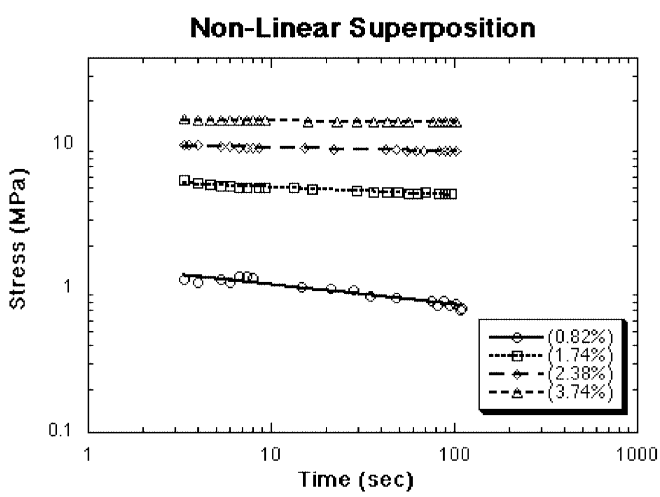

Fig. 3. Application of single integral modified (nonlinear) Superposition theory (*lines*) to experimental stress-relaxation results of Provenzano et al. (2001) (*points*) for multiple testing at various strain levels of a single rat ligament. R^2 = 0.91, 0.91, 0.96, and 0.95 for ε = 0.82%, 1.74%, 2.38%, and 3.74%, respectively

A polynomial describing the rate function, $B(\varepsilon) = -0.1902 + 0.1364\varepsilon - 0.03403\varepsilon^2 + 0.002765\varepsilon^3$; $R^2 = 0.92$, was obtained by fitting rate versus strain data from Provenzano et al. (2001) (Fig. 4). Three separate stress-relaxation tests were then fitted using the predicted form of Eq. (11) (Fig. 5). Using the predicted rate formulation, the curves were fit with R^2 values of 0.84, 0.78, and 0.99 for strains of 0.85, 2.20, and 2.47%, respectively. Fitting each curve individually with the modified superposition formulation resulted in R^2 values of 0.97, 0.99, and 0.99 for strains of 0.85, 2.20, and 2.47%, respectively. Hence, as expected, scatter in the experimental data limits the predicted rate formulation. However, the prediction formulation fit the curves reasonably well and shows promise as a predictive measure of stress relaxation.

Behavior of the modied superposition method under sinusoidal strain history

The strain-dependent function $A(\varepsilon)$ and $B(\varepsilon)$ in Eq. (13) were obtained from experimental rat MCL data. The function $A(\varepsilon)$ was determined by fitting the tangential modulus as a function of strain from typical stress–strain data for the rat MCL: $A(\varepsilon) = 3636.7\varepsilon^{0.7173}$, $R^2 > 0.97$. The function $B(\varepsilon)$ was chosen to be the same as described in the previous section (Fig. 4): $B(\varepsilon) = -0.1902 + 0.1364\varepsilon - 0.03403\varepsilon^2 + 0.002765\varepsilon^3$; $R^2 = 0.92$. Figure 6a displays model behavior over 25 cycles at peak strain (ε_0) levels of 0.5 and 1.5%, for a frequency factor (α) equal to one. The peak stresses at a constant frequency factor display strain-dependent behavior, although to a lesser extent than seen with constant strain (Fig. 6b). The magnitude of stress varies with frequency (Fig. 7). When examining model behavior over one order of magnitude of frequency ($\alpha = 0.1$ to 1.0), initial stress increases from 1.27 to 1.95 MPa. In addition, valley stresses increase with increasing frequency. Relaxation rate, determined by fitting peak stresses with a power law, increases with increasing frequency: –0.07 at $\alpha = 0.1$ to –0.13 at $\alpha = 1.0$. Hence, the model predicts that the relaxation rate is both strain and frequency dependent.

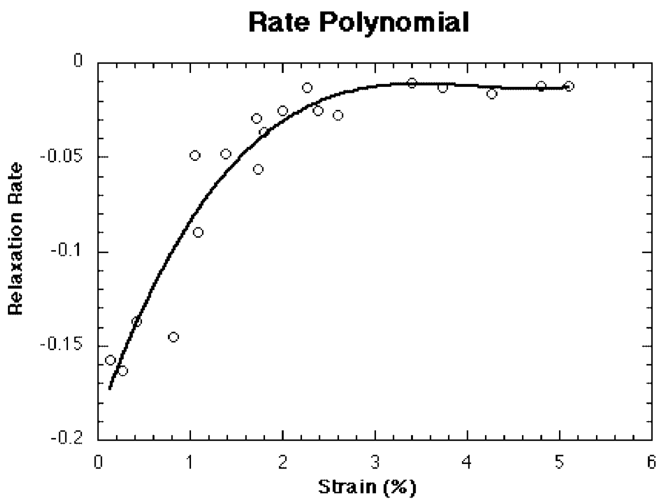

Fig. 4. Fitting of a polynomial function (*curve*) to experimental stress-relaxation rate of Provenzano et al. (2001) (*points*) for multiple rat ligaments tested at multiple strain levels. Rate was defined as n in a t^n time dependence. The strain-dependent rate behavior in this study can be described by a polynomial function for $B(\varepsilon)$ as: $B(\varepsilon) = -0.1902 + 0.1364\,\varepsilon - 0.03403\varepsilon^2 + 0.002765\varepsilon^3$; $R^2 = 0.92$

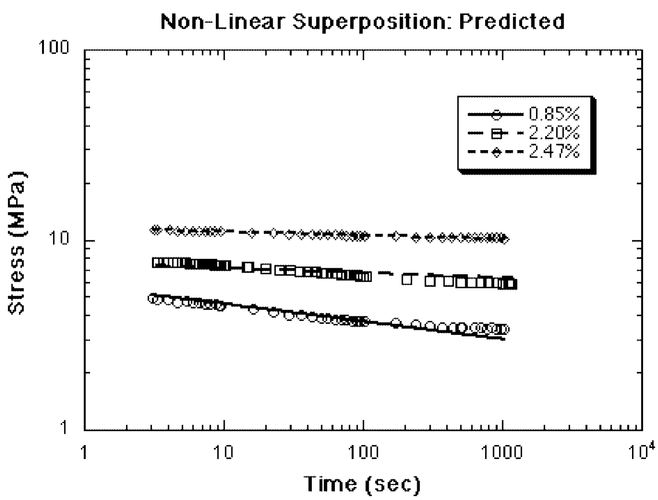

Fig. 5. Comparison of experimental data (*points*) and predicted (*lines*) stress-relaxation behavior. Predictions are based on curve fitting of a prior set of stress-relaxation data (Fig. 3) at different strain levels. $R^2 = 0.84$, 0.78, and 0.99 for strains of 0.85, 2.20, and 2.47%, respectively

4
Discussion

This study investigated the ability of two nonlinear viscoelastic models to describe experimentally observed ligament behavior. Models in which the time-dependent behavior is independent of stress or strain cannot adequately describe the reported nonlinear behavior of ligament. The two nonlinear theories examined in this study described both elastic and viscous nonlinear behavior well.

The utility of phenomenological models, such as the ones employed in this study, lies in the fact that experimental results can be obtained only over a discrete region of the independent variables, in this case time and strain. Models provide parameters that can be used to compare experimental data sets and allow one to predict the behavior in intermediate regions not covered by the experiments. For such an approach to succeed, the dependence must satisfy conditions of smoothness. Although sharp peaks and other abrupt behavior are known in resonating systems, they do not occur in systems that relax. Therefore the approach is warranted. Phenomenological models can also be used in finite element analyses to predict

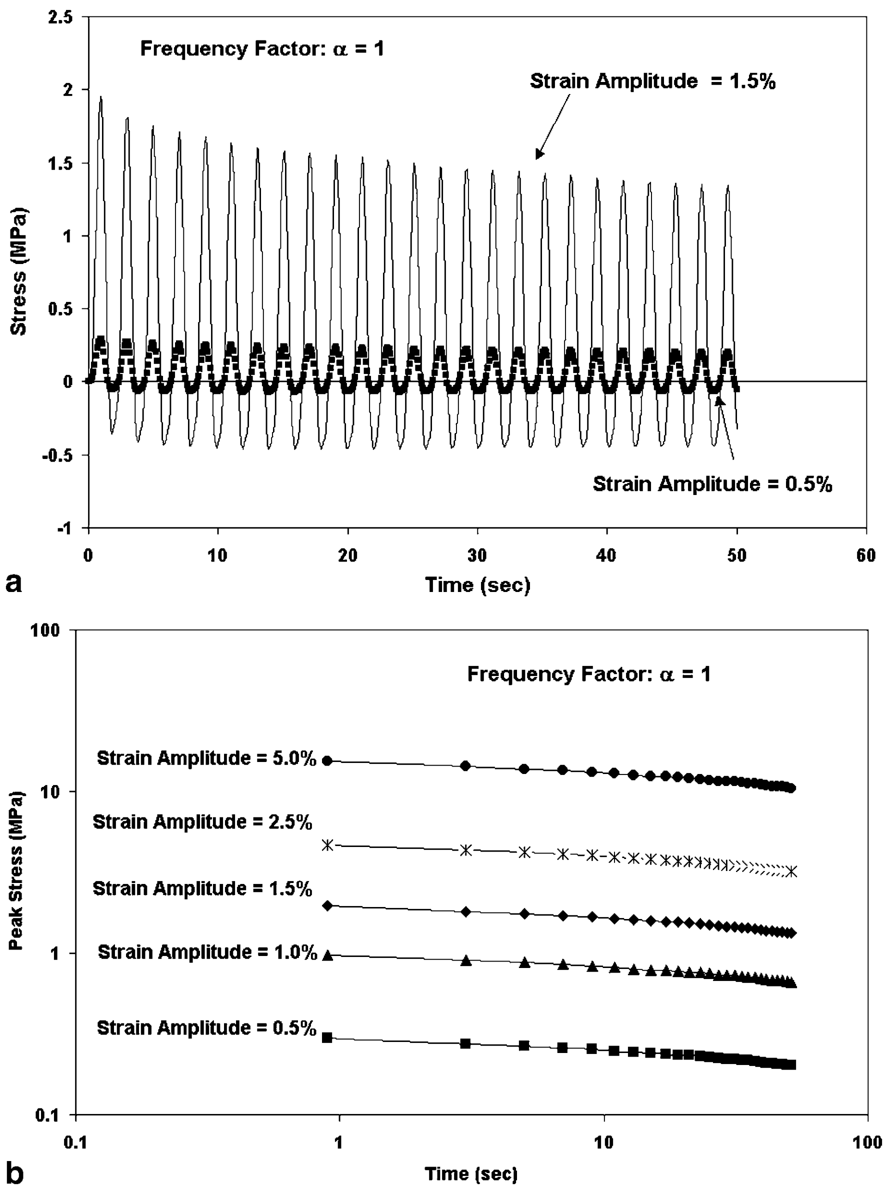

Fig. 6. **a** Sinusoidal stress behavior at strain amplitude of 0.5 and 1.5% over 25 cycles for $\alpha = 1$. **b** Peak stresses for 0.5, 1.0, 1.5, 2.5, and 5.0% peak strains with $\alpha = 1$. The rate of relaxation of the peak stresses shows strain-dependent behavior: $\varepsilon_0 = 0.5\%$, rate = –0.1014; $\varepsilon_0 = 1.0\%$, rate = –0.1012; $\varepsilon_0 = 1.5\%$, rate = –0.1010; $\varepsilon_0 = 2.5\%$, rate = –0.1006; $\varepsilon_0 = 5.0\%$, rate = –0.0996

stress and strain distributions in tissues having complex geometries or subjected to complex loads.

Both Schapery's theory and modified superposition are able to successfully model the data examined thus far. This is perhaps not surprising since these formulations have some similarities at constant temperature, though the Schapery form offers more flexibility in the number of variable characteristic functions. However, in this application of Schapery's theory not all the available freedom of the model was needed. For instance, Schapery's theory can have upward of six unknown functions and constants (h_e, h_1, h_2, a_e, and terms of the transient modulus) allowing the model to be adaptable. In this application the Schapery model was reduced to two unknown functions of strain (h_e and h_2) and two unknown constants C and n, thus not all the flexibility of the theory was required. From Schapery's theory, information regarding the initial, final, and relaxation modulus can be easily obtained, as can an indication of the degree of nonlinearity by examining the h terms. In this application, the decrease in h_2 with increasing strain indicates a decrease in the rate of relaxation with increasing strain. Modified superposition allows the initial (isochronal) modulus or stress to be easily obtained as well as an indication of the degree of rate dependence on strain through the exponent of the power law.

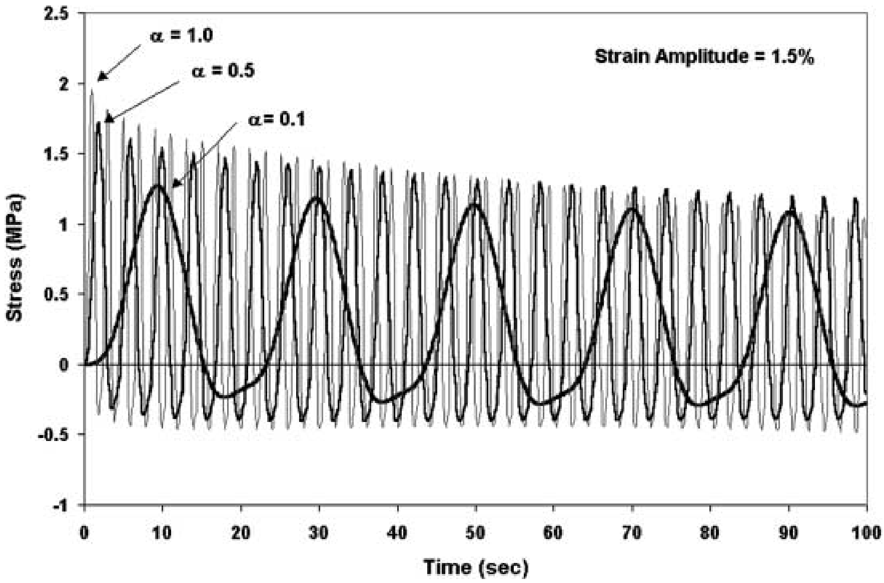

Fig. 7. Sinusoidal stress behavior at $\alpha = 0.1$, 0.5, and 1.0 for $\varepsilon_0 = 1.5\%$. Peak and valley stress-display frequency dependent behavior with larger initial stresses associated with faster loading. Relaxation rate, determined by fitting peak stresses with a power law, increased with increasing frequency: $\alpha = 0.1$, rate = –0.0695; $\alpha = 0.5$, rate = –0.1005; $\alpha = 1.0$, rate = –0.1325

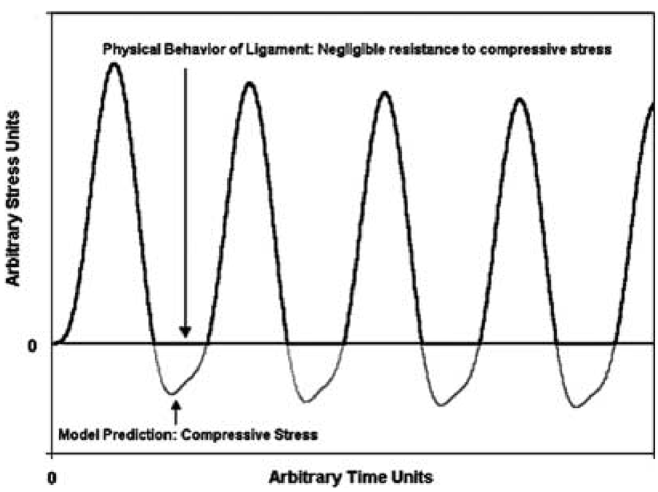

Fig. 8. Example of physical ligament behavior. Medial collateral ligament does not substantially resist compressive loading. Therefore, as the tissue is brought back to zero strain, the tissue stress will appear to plateau (*black lines*). Equation (13) predicts compressive stress in the tissue (*gray lines*). This will only be physically accurate if tissues were capable of resisting compressive loading

Since the modulus formulation used with modified superposition in this study allows the rate dependent nonlinearity to be accounted for with one term, the degree of nonlinearity can be easily determined and future predictive formulations (such as shown above) are more easily obtained. For this reason, modified superposition allows for a more direct interpretation of the relationship between model parameters and physical behavior than does Schapery's theory.

A sinusoidal strain history was applied within the framework of the modified superposition formulation. The model predicted that the rate of stress relaxation is both strain and frequency dependent. Peak and valley stresses increased with strain and frequency. Woo et al. (1990) examined the effect of strain rate on material properties of rabbit MCLs. Despite differences between their experiments and simulations in this study, the qualitative differences in initial peak stresses over one order of magnitude ($\sim$0.15–1.5%/s) appear consistent. In regard to compressive stresses, it is clear that ligament does not resist the compressive loads predicted by the model. A more realistic plot of a ligament's response to a haversine tensile stretch is shown in Fig. 8, in which stress does not go below zero, but instead plateaus until tensile loading resumes. With this load history, the rate of relaxation (of peak stresses) was dependent upon strain (ε_0), but not to the degree seen experimentally under constant strain. The recovery period inherent in sinusoidal loading is a likely cause for the reduction in strain-dependent behavior and the rate of recovery is likely to also be strain dependent. In addition, the relaxation rate nonlinearities seen in step loadings are attenuated by their superposition in the Boltzmann integral with this sinusoidal load history. However, further experimental studies need to be performed before the theoretical behavior presented can be confirmed and discussed in detail.

The mechanisms driving viscoelastic behavior in ligament are not yet completely defined. It has been speculated that "the decrease in relaxation rate with increasing strain could be the result of larger strains causing greater water loss (wringing out effect) which causes the tissue to be more elastic (less viscous) than tissues subjected to lower strains" (Provenzano et al. 2001). Studies supporting this hypothesis have reported increased relaxation with increased hydration (Chimich et al. 1992) and a decrease in tissue water content with cyclic loading (Hannafin and Arnoczky 1994). Thornton et al. (1997) speculated that creep behavior is due to the progressive recruitment of collagen fibers during creep (Thornton et al. 2001) and that this microstructural behavior is unlikely to have as substantial an effect on stress relaxation as on creep. It has been hypothesized that this behavior could also explain the decrease in the rate of creep with increasing load (Provenzano et al. 2001). As larger forces are applied to the ligament, more fibers are recruited, leaving fewer fibers to be progressively recruited after initial loading, which would therefore decrease the creep response. If these mechanisms are correct, then the differences in the rate behavior with stress or strain would have thermodynamic and/or structural significance. For instance, a change in stress-relaxation rate behavior caused by changes in hydration, or changes in creep behavior caused by changes in the degree of progressive fiber recruitment, may be considered in the context of irreversible thermodynamics or using structural models, such as the model by Hurschler et al. (1997), which incorporates collagen fiber recruitment. Yet, further understanding of the physical mechanisms driving nonlinear viscoelastic behavior in ligament are required before the physical significance of strain-dependent rate terms, such as $B(\varepsilon)$, can be interpreted.

Several limitations must be borne in mind when considering the models examined in this study. The data examined do not identify all possible nonlinearities in ligament, such as the effects of age, healing, biochemical changes, temperature, hydration, and others. In addition, only single integral formulations are examined. Single integral formulations are more manageable than multiple integral formulations such as that of Green and Rivlin (1957). Using a 1-D single integral formulation, only a subset of viscoelastic phenomena can be described, while multiple integral or tensorial formulations are more robust and allow for more complex behavior such as multi-axial states of stress (Findley et al. 1976). Second, the experimental rat MCL data (Provenzano et al. 2001) examined in this study are limited and to the authors knowledge only one other study (Haridas et al. 2001) identifies strain-dependent rate behavior in tendon. A larger range of strains for stress relaxation or stresses for creep and longer times (over many decades) needs to be examined to identify relevant nonlinearities. In addition, extremes of loading rate have not been examined. Experimental studies under sinusoidal strain need to be performed in order to support the behavior indicated by modified superposition under a sinusoidal strain history. Finally, studies still have not explored all aspects of ligament viscoelastic behavior during various human activities, which may include complex creep and relaxation components.

QLV has been, and remains, a valuable tool in the field of biomechanics. However, the QLV formulation cannot describe all viscoelastic behavior and for such data more general formulations are required. Two such formulations were considered in this study, Schapery's theory (1969) and modified superposition (Findley et al. 1976), both of which describe the reported (Provenzano et al. 2001) nonlinear viscoelastic behavior well. However, modified superposition allows for easier interpretation of elastic and rate nonlinearity when using a power law formulation since the rate is revealed in the exponent of the power law, $B(\varepsilon)$, and the amplitude term, $A(\varepsilon)$, reveals elastic nonlinearity. Further work remains with respect to interrelating stress-relaxation and creep data with strain- or stress-dependent time behavior, respectively.

References

Atkinson, T.S.; Ewers, B.J.; Haut, R.C.: The tensile and stress-relaxation responses of human patellar tendon varies with specimen cross-sectional area. J Biomech 32 (1999) 907–914

Bausch, A.R.; Moller, W.; Sackmann, E.: Measurement of local viscoelasticity and forces in living cells by magnetic tweezers. Biophys J 76 (1999) 573–579

Best, T.M.; McElhaney, J.; Garrett, W.E.; Myers, B.S.: Characterization of the passive responses of live skeletal muscle using the quasi-linear theory of viscoelasticity. J Biomech 27 (1994) 413–419

Bingham, D.N.; DeHoff, P.H.: A constitutive equation for the canine anterior cruciate ligament. J Biomech Eng 101 (1979) 15–22

Carew, E.O.; Barber, J.E.; Vesely, I.: Role of preconditioning and recovery time in repeated testing of aortic valve tissues: validation through quasilinear viscoelastic theory. Ann Biomed Eng 28 (2000) 1093–1100

Carew, E.O.; Talman, E.A.; Boughner, D.R.; Vesely, I.: Quasi-linear viscoelastic theory applied to internal shearing of porcine aortic valve leaflets. J Biomech Eng 121 (1999) 386–392

Chimich, D.; Shrive, N.; Frank, C.; Marchuk, L.; Bray, R.: Water content alters viscoelastic behavior of the normal adolescent rabbit medial collateral ligament. J Biomech 25 (1992) 831–837

Christensen, R.M.: A nonlinear theory of viscoelasticity for application to elastomers. J Appl Mech 47 (1980) 762–768

Corr, D.T.; Strarr, M.J.; Vanderby, R. Jr.; Best, T.M.: A nonlinear generalized Maxwell fluid model for viscoelastic materials. J Appl Mech 68 (2001) 787–790

Decraemer, W.F.; Maes, M.A.; Vanhuyse, V.J.; Vanpeperstraete, P.: A non-linear viscoelastic constitutive equation for soft biological tissues, based upon a structural model. J Biomech 13 (1980) 559–564

Dehoff, P.H.: On the nonlinear viscoelastic behavior of soft biological tissues. J Biomech 11 (1978) 35–40

Dillard, D.A.; Straight, M.R.; Brinson, H.F.: The nonlinear viscoelastic characterization of graphite/epoxy composites. Polymer Eng Sci 27 (1987) 116–123

Egan, J.M.: A constitutive model for the mechanical behaviour of soft connective tissues. J Biomech 20 (1987) 681–692

Findley, W.N.; Lai, J.S.; Onaran, K.: Creep and relaxation of nonlinear viscoelastic materials. Dover, New York, (1976)

Fung, Y.C.: Stress–strain history relations of soft tissues in simple elongation. In: Fung YC, Perrone N, Anliker M (eds) Stress–strain history relations of soft tissues in simple elongation. Prentice Hall, Englewood Cliffs, N.J., (1972)

Graf, B.K.; Vanderby, R. Jr.; Ulm, M.J.; Rogalski, R.P.; Thielke, R.J.: Effect of preconditioning on the viscoelastic response of primate patellar tendon. Arthroscopy 10 (1994) 90–96

Green, E.; Rivlin, R.S.: The mechanics of non-linear materials with memory. Arch Ration Mech Anal 1 (1957) 1–21

Guilak, F.: The deformation behavior and viscoelastic properties of chondrocytes in articular cartilage. Biorheology 37 (2000) 27–44

Guilak, F.; Tedrow, J.R.; Burgkart, R.: Viscoelastic properties of the cell nucleus. Biochem Biophys Res Commun 269 (2000) 781–786

Guilak, F.; Ting-Beall, H.P.; Baer, A.E.; Trickey, W.R.; Erickson, G.R.; Setton, L.A.: Viscoelastic properties of intervertebral disc cells. Identification of two biomechanically distinct cell populations. Spine 24 (1999) 2475–2483

Hannafin, J.A.; Arnoczky, S.P.: Effect of cyclic and static tensile loading on water content and solute diffusion in canine flexor tendons: an in vitro study. J Orthop Res 12 (1994) 350–356

Haridas, B.; Butler, D.L.; Weiss, J.A.: Age dependent differences in compressive viscoelasticity in the rabbit flexor tendon. In: Proceedings of the ASME Bioengineering Conference, Snowbird, Utah, vol 50. ASME, New York, p 105 (2001)

Haut, R.C.; Little, R.W.: Rheological properties of canine anterior cruciate ligaments. J Biomech 2 (1969) 289–298

Heidemann, S.R.; Kaech, S.; Buxbaum, R.E.; Matus, A.: Direct observations of the mechanical behaviors of the cytoskeleton in living fibroblasts. J Cell Biol 145 (1999) 109–122

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

Johnson, G.A.; Livesay, G.A.; Woo, S.L.; Rajagopal, K.R.: A single integral finite strain viscoelastic model of ligaments and tendons. J Biomech Eng 118 (1996) 221–226

Lai, J.S.Y.; Findley, W.N.: Stress relaxation of nonlinear viscoelastic material under uniaxial strain. Trans Soc Rheol 12 (1968) 259–280

Lakes, R.S.: Viscoelastic solids. CRC Press, New York, (1998)

Lakes, R.S.; Katz, J.L.: Viscoelastic properties of wet cortical bone – III. A non-linear constitutive equation. J Biomech 12 (1979) 689–698

Lakes, R.S.; Katz, J.L.; Sternstein, S.S.: Viscoelastic properties of wet cortical bone – I. Torsional and biaxial studies. J Biomech 12 (1979) 657–678

Lakes, R.S.; Vanderby, R.: Interrelation of creep and relaxation: a modeling approach for ligaments. J Biomech Eng 121 (1999) 612–615

Lanir, Y.: The rheological behavior of the skin: experimental results and a structural model. Biorheology 16 (1979) 191–202

Lanir, Y.: A microstructure model for the rheology of mammalian tendon. J Biomech Eng 102 (1980) 332–339

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

Lou, Y.C.; Schapery, R.A.: Viscoelastic characterization of a nonlinear fiber-reinforced plastic. J Composite Mater 5 (1971) 208–234

Mak, A.F.: The apparent viscoelastic behavior of articular cartilage – the contributions from the intrinsic matrix viscoelasticity and interstitial fluid flows. J Biomech Eng 108 (1986) 123–130

Pipkin, A.C.; Rogers, T.G.: A nonlinear integral representation for viscoelastic behavior. J Mech Phys Solids 16 (1968) 59–74

Provenzano, P.P.; Heisey, D.; Haysashi, K.; Lakes, R.S.; Vanderby, R. Jr.: Sub-failure damage in ligament: a structural and cellular evaluation. J Appl Physiol 92 (2002) 362–371

Provenzano, P.P.; Lakes, R.S.; Keenan, T.; Vanderby, R.J.: Non-linear ligament viscoelasticity. Ann Biomed Eng 29 (2001) 908–914

Rousseau, E.P.; Sauren, A.A.; van Hout, M.C.; van Steenhoven, A.A.: Elastic and viscoelastic material behaviour of fresh and glutaraldehyde-treated porcine aortic valve tissue. J Biomech 16 (1983) 339–348

Sanjeevi, R.; Somanathan, N.; Ramaswamy, D.: A viscoelastic model for collagen fibres. J Biomech 15 (1982) 181–183

Sauren, A.A.; Rousseau, E.P.: A concise sensitivity analysis of the quasi-linear viscoelastic model proposed by Fung. J Biomech Eng 105 (1983) 92–95

Sauren, A.A.; van Hout, M.C.; van Steenhoven, A.A.; Veldpaus, F.E.; Janssen, J.D.: The mechanical properties of porcine aortic valve tissues. J Biomech 16 (1983) 327–337

Schapery, R.A.: A theory of nonlinear thermoviscoelasticity based on irreversible thermodynamics. In: Proceedings of the 5th U.S. National Congress on Applied mechanics. ASME, New York, p 511 (1966)

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

Shoemaker, P.A.; Schneider, D.; Lee, M.C.; Fung, Y.C.: A constitutive model for two-dimensional soft tissues and its application to experimental data. J Biomech 19 (1986) 695–702

Thornton, G.M.; Frank, C.B.; Shrive, N.G.: Ligament creep behavior can be predicted from stress relaxation by incorporating fiber recruitment. J Rheol 45 (2001) 493–507

Thornton, G.M.; Oliynyk, A.; Frank, C.B.; Shrive, N.G.: Ligament creep cannot be predicted from stress relaxation at low stress: a biomechanical study of the rabbit medial collateral ligament. J Orthop Res 15 (1997) 652–656

Touti, D.; Cederbaum, G.: On the prediction of stress relaxation from known creep in nonlinear materials. J Eng Mat Tech 119 (1997) 121–124

Trickey, T.R.; Lee, M.; Guilak. T.: Viscoelastic properties of chondrocytes from normal and osteoarthritic human cartilage. J Orthop Res 18 (2000) 891–898

Viidik, A.: A rheological model for uncalcified parallel-fibered collagenous tissue. J Biomech 1 (1968) 3–11

Woo, S.L.: Mechanical properties of tendons and ligaments – I. Quasi-static and nonlinear viscoelastic properties. Biorheology 19 (1982) 385–396

Woo, S.L.; Gomez, M.A.; Akeson, W.H.: The time and history-dependent viscoelastic properties of the canine medical collateral ligament. J Biomech Eng 103 (1981) 293–298

Woo, S.L.; Peterson, R.H.; Ohland, K.J.; Sites, T.J.; Danto, M.I.: The effects of strain rate on the properties of the medial collateral ligament in skeletally immature and mature rabbits: a biomechanical and histological study. J Orthop Res 8 (1990) 712–721

Woo, S.L.; Simon, B.R.; Kuei, S.C.; Akeson, W.H.: Quasi-linear viscoelastic properties of normal articular cartilage. J Biomech Eng 102 (1980) 85–90