Acta of Bioengineering and Biomechanics
Vol. 16, No. 4, 2014

Original paper
DOI: 10.5277/ABB-00005-2014-03

Non-integer viscoelastic constitutive law to model soft biological tissues to in-vivo indentation

NAGEHAN DEMİRCİ[1], ERGİN TÖNÜK[2]*

[1] Department of Mechanical Engineering, Middle East Technical University, Ankara, Turkey.
[2] Department of Mechanical Engineering, Graduate Program of Biomedical Engineering, Biomaten,
Center of Exellence in Biomaterials and Tissue Engineering, Middle East Technical University, 06800 Ankara, Turkey.

Purpose: During the last decades, derivatives and integrals of non-integer orders are being more commonly used for the description of constitutive behavior of various viscoelastic materials including soft biological tissues. Compared to integer order constitutive relations, non-integer order viscoelastic material models of soft biological tissues are capable of capturing a wider range of viscoelastic behavior obtained from experiments. Although integer order models may yield comparably accurate results, non-integer order material models have less number of parameters to be identified in addition to description of an intermediate material that can monotonically and continuously be adjusted in between an ideal elastic solid and an ideal viscous fluid.

Methods: In this work, starting with some preliminaries on non-integer (fractional) calculus, the "spring-pot", (intermediate mechanical element between a solid and a fluid), non-integer order three element (Zener) solid model, finally a user-defined large strain non-integer order viscoelastic constitutive model was constructed to be used in finite element simulations. Using the constitutive equation developed, by utilizing inverse finite element method and *in vivo* indentation experiments, soft tissue material identification was performed.

Results: The results indicate that material coefficients obtained from relaxation experiments, when optimized with creep experimental data could simulate relaxation, creep and cyclic loading and unloading experiments accurately.

Conclusions: Non-integer calculus viscoelastic constitutive models, having physical interpretation and modeling experimental data accurately is a good alternative to classical phenomenological viscoelastic constitutive equations.

Key words: *fractional calculus, indentation tests, inverse finite element analysis, soft tissue constitutive relation, viscoelasticity*

1. Introduction

The mathematics of "fractional" calculus dates back to the 17th century when Leibniz introduces the notation $D^n y = \frac{d^n y}{dx^n}$ for differentiation where n is a non-negative integer, and in 1695, L'Hospital in a letter to Leibniz asks what happens if the order of derivative, n, becomes ½? The response of Leibniz is interesting: "It is a paradox but some day useful consequences will be drawn" [29]. The non-integer order (commonly used word fractional is a misnomer) differential and integral calculus may take not only the fractional numbers, but it may include any arbitrary number as the order of differentiation or integration. As Leibniz predicted, this property of calculus becomes a powerful tool recently in various science and engineering applications. The theory of non-integer order calculus is thoroughly reviewed in many references, e.g., Ross [29], Oldham and Spanier [22], Podlubny [25], Bayın [1], Dalir and Bashour [7], Machado et al. [20].

The most popular expression of the non-integer order integration in notation of Davis [8] is the Riemann–Liouville non-integer order integral which is also called the "integro-differential" expression (equation 1).

$$ {}_aD_x^{-\beta} f(x) = \frac{1}{\Gamma(\beta)} \int_a^x (x-t)^{\beta-1} f(t)dt, \quad \mathrm{Re}(\beta) > 0 \,. \quad (1)$$

The subscripts a and x denote the limits of the integration, called the "terminals" and $\Gamma(\beta)$ is the

* Corresponding author: Orta Doğu Teknik Üniversitesi Makina Mühendesliği Bölümü, Üniversiteler Mahallesi, Dumlupınar Bulvarı, 06800 Ankara, Turkey. Tel: 903122105293, e-mail: tonuk@metu.edu.tr
Received: January 31st, 2014
Accepted for publication: May 26th, 2014

Euler's gamma function, which generalizes the factorial, $\beta!$, defined only for integers, to real and complex arguments by $\Gamma(\beta) = (\beta - 1)!$.

The non-integer order derivative is not only dependent on the value of the function at the point considered (and within infinitesimal neighborhood) but the value of the function on the whole interval which assures 'non-locality' of the integro-differential operator. Therefore, unlike ordinary derivatives, which are point functionals, non-integer order derivatives are hereditary functionals possessing total memory of the past states [11] which is a better candidate for the description of memory and hereditary properties of viscoelastic materials, including soft biological tissues.

The history of non-integer order calculus on the theory of viscoelasticity dates back to the Nutting's [21] observations in 1921 on the stress relaxation phenomena which is described by fractional powers of time than decaying exponentials. Gemant [16] proposes a viscoelastic model with a derivative order of less than one. Scott-Blair [30], [32], [33] obtains the non-integer order differential expression for "intermediate" materials being neither fluid nor solid. Rabotnov [28] uses Volterra integral operators to define constitutive equation of a viscoelastic material. Considerable work devoted to one-dimensional non-integer order viscoelastic material models exist in literature including some soft biological tissue applications. Caputo and Mainardi [2], [3] obtain the creep and relaxation functions in terms of the one-term Mittag–Leffler function by using the Laplace transform and Koeller [18] generalizes some of the concepts of Rabotnov's theory to demonstrate the relation between non-integer calculus and the theory of linear viscoelasticity, and derives the creep and relaxation functions for the non-integer order Maxwell, Kelvin–Voigt and three-element (Zener) material models by employing Rabotnov's Theory of Hereditary Solid Mechanics [28]. These developments in the theoretical aspect of non-integer calculus makes it possible to use non-integer order viscoelasticity for material models, including biomechanics. Suki et al. [34] employ three-parameter non-integer order equations for the description of pressure-volume response of the rat whole lung. Djordjevic et al. [12] employ non-integer calculus methods to define the smooth muscle cell rheological behavior. Doehring et al. [13] augment a non-integer order relaxation function to Fung's quasi-linear viscoelasticity (QLV) theory to describe the aortic valve cusp biomechanics. Craiem and Armentano [6] analyze the arterial wall mechanics by employing a four parameter fractional order material model. Kohandel et al. [19] and Davis et al. [8] compare the integer and non-integer order constitutive equations of brain parenchyma. Freed and Diethelm [14] extend the elastic law of Fung, which describes the nonlinear stress-strain behavior of soft biological tissues, to a viscoelastic material model which includes non-integer order derivatives of Caputo type. Craiem et al. [5] investigate the arterial wall mechanics by employing fractional operators to Fung's reduced relaxation function. Grahovac and Zigic [17] employ non-integer order derivatives for modeling the hamstring muscle group in ramp-and-hold stress relaxation. Wilkie et al. [36] formulate a hyperelastic, fractional viscoelastic model to model the infant brain tissue. Chang et al. [4] analyze the impact response of human head frontal bone, where the soft tissues surrounding the frontal bone are simulated by a fractional derivative standard linear solid (Zener) model.

In this work, starting with a so-called "*spring-pot*", an intermediate mechanical element between an ideal elastic spring and an ideal viscous dashpot, the constitutive equations for a non-integer order three element (Zener) model and finite strain non-integer order viscoelastic constitutive equation to be used in a commercial finite element software (Msc. Marc-Mentat® 2010) are presented. The non-integer order three element viscoelastic material model was tested by in vivo indentation experiments. Inverse finite element method was used to identify the material constants of finite strain non-integer order material model that simulates in vivo indentation experiments presented by Petekkaya [23] and Petekkaya and Tönük [24].

2. Materials and methods

In classical integer order viscoelastic modeling, the mechanical behavior is modeled by a physical mixture of elastic solid(s) and Newtonian viscous fluid(s). In the simplest one-dimensional material models the ideal elastic spring represents the Hookean elastic solid and the dashpot element represents the Newtonian viscous fluid. The phenomenological material models are developed by connecting a finite number of these mechanical elements in series or in parallel to obtain the desired viscoelastic response observed through experiments. On the other hand, non-integer order viscoelastic models have an intermediate element between a linear elastic spring and a viscous dashpot termed as the *spring-pot*.

The spring-pot

The constitutive equation for a spring-pot is [31] presented in equation (2).

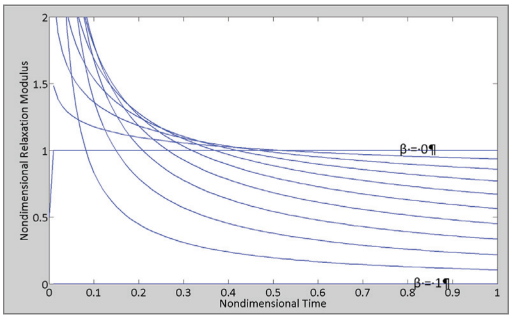
$$\sigma(t) = E\eta^{\beta}\frac{d^{\beta}\varepsilon}{dt^{\beta}}, \quad 0 \le \beta \le 1. \tag{2}$$

Here η is the characteristic time and non-integer order, β is the memory parameter characterizing the power law response. For $0 \le \beta \le 1$ the spring-pot element provides a smooth, continuous and monotonic transition from elastic ($\beta = 0$) to viscous ($\beta = 1$) response. Relaxation modulus and creep compliance of a spring-pot are presented in equations (3) and (4), respectively.

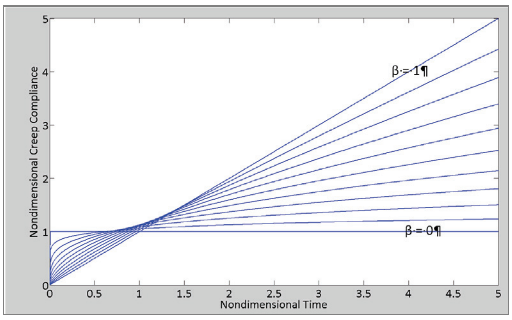
$$G(t) = \frac{E}{\Gamma(1-\beta)}\left(\frac{t}{\eta}\right)^{-\beta}, \tag{3}$$

$$J(t) = \frac{1}{E\Gamma(1+\beta)}\left(\frac{t}{\eta}\right)^{\beta}. \tag{4}$$

For different values of the memory parameter, β, the normalized relaxation modulus and creep compliance versus non-dimensional time, t/τ, of the spring-pot is presented in Fig. 1, where the monotonic transition from elastic to viscous response is clearly observed.



(a) Relaxation



(b) Creep

Fig. 1. Non-dimensional relaxation modulus and creep compliance versus non-dimensional time for the spring-pot for the non-integer parameter β from 0 to 1 in steps of 0.1 a) relaxation, b) creep

When the dashpot in the three-element (Zener) viscoelastic model is replaced by a spring-pot, (Fig. 2) the governing equation for the resulting mechanical model is presented in equation (5). The relaxation modulus and creep compliance of the non-integer order three-element model are presented in equations (6) and (7), respectively.

$$\left(\frac{d^{\beta}}{dt^{\beta}} + \frac{E_1 + E_2}{E_2\eta^{\beta}}\right)\sigma(t) = E_1\left(\frac{d^{\beta}}{dt^{\beta}} + \frac{1}{\eta^{\beta}}\right)\varepsilon(t), \tag{5}$$

$$G(t) = E_1\left\{1 - \frac{E_1/E_2}{1+E_1/E_2}\left\{1 - E_{\beta}\left[-\left(\frac{t}{\tau}\right)^{\beta}\right]\right\}\right\}, \tag{6}$$

$$J(t) = \frac{1}{E_1}\left\{1 + \frac{E_1}{E_2}\left\{1 - E_{\beta}\left[-\left(\frac{t}{\tau}\right)^{\beta}\right]\right\}\right\}. \tag{7}$$

In these equations, $\tau = \frac{\eta}{\sqrt[\beta]{1+E_1/E_2}}$, $\eta = \frac{F}{E_2}$ and $E_{\beta}(\cdot)$ is the one parameter Mittag–Leffler function. E_2 and F are the elastic modulus and viscosity of the spring-pot for the limiting values of $\beta = 0$ and $\beta = 1$, respectively.

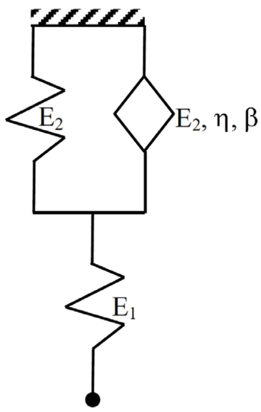
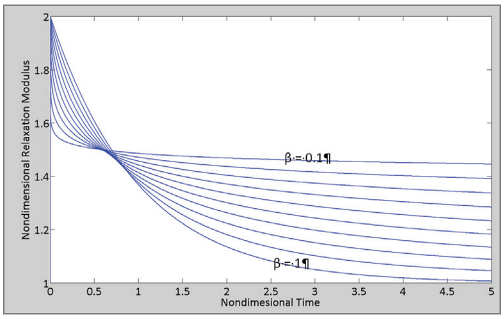


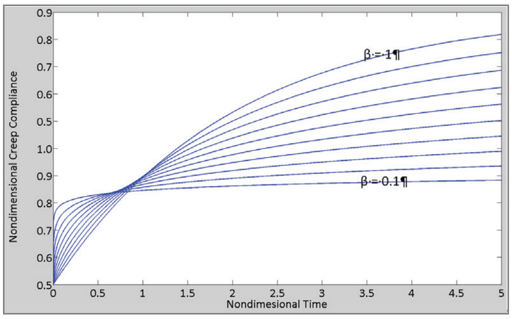
Fig. 2. Three element non-integer standard linear solid model

The relaxation modulus and creep compliance versus non-dimensional time of the non-integer order three element standard linear solid for different values of non-integer order β are presented in Fig. 3.

The fading memory of the model becomes more dominant as non-integer order parameter β, decreases. E_1 and E_2 determine the magnitudes of initial and final plateaus and time constant τ determines the rate of relaxation and creep.



(a) Relaxation



(b) Creep

Fig. 3. Non-dimensional relaxation modulus and creep compliance versus non-dimensional time for $E_1/E_2 = 1$ for the non-integer parameter β from 0.1 to 1 in steps of 0.1 a) relaxation, b) creep

Mittag–Leffler function

The Mittag–Leffler function is the "generalized" (natural expansions of the) exponential function and appears in the analytic solutions of the non-integer order differential equations similar to exponential functions seen in solutions of integer order differential equations.

The two-parameter Mittag–Leffler function (equation (8)) is a completely monotonic function in power series representation for $0 < \beta \leq 1$. The one-parameter Mittag-Leffler function is obtained by setting the second parameter (β) to 1 and replacing the first parameter (α) by β (equation (9)). Mittag–Leffler function boils down to exponential function if both parameters in two parameter function are set to unity or the parameter in one parameter Mittag–Leffler function is set to unity (equation (10)).

$$E_{\alpha,\beta}(x) = \sum_{k=0}^{\infty} \frac{x^k}{\Gamma(\alpha k + \beta)}, \quad \alpha > 0, \beta > 0, \quad (8)$$

$$E_{\beta}(x) = E_{\beta,1}(x) \sum_{k=0}^{\infty} \frac{x^k}{\Gamma(\beta k + 1)}, \quad \beta > 0, \quad (9)$$

$$E_{1,1}(x) = E_1(x) = \exp(x) = e^x. \quad (10)$$

Diethelm et al. [11] proposed a fast computational scheme for the evaluation of one parameter Mittag–Leffler function based on Padé approximation applied for different intervals as presented in equation (11). The coefficients, a_i and b_i, are tabulated in the original publication.

$$E_\beta(-x^\beta) \simeq \begin{cases} -\sum_{k=0}^{4} \frac{(-x)^{\beta k}}{\Gamma(1+\beta k)}, & 0 \leq x \leq 0.1, \\ \frac{a_0 + a_1 x + a_2 x^2}{1 + b_1 x + b_2 x^2 + b_3 x^3}, & 0.1 < x < 15, \\ -\sum_{k=0}^{4} \frac{x^{-\beta k}}{\Gamma(1-\beta k)}, & x \geq 15. \end{cases} \quad (11)$$

Finite strain non-integer order viscoelastic material model

The instantaneous elastic and time dependent non-integer time derivative parts of the soft tissue strain energy density function relating Green–Lagrange finite strain tensor to second Piola–Kirchhoff stress tensor was modeled to be multiplicatively separable as presented in equation (12).

$$W(C,t) = W_0(C)G(t) \quad (12)$$

where W_0 is the instantaneous (glassy) strain energy density function for a hyper-elastic material modeled with five phenomenological material constants in the James–Green–Simpson form (equation (13)).

$$W_0 = C_{10}(I_1 - 3) + C_{01}(I_2 - 3) + C_{11}(I_1 - 3)(I_2 - 3) + C_{20}(I_1 - 3)^2 + C_{30}(I_1 - 3)^3. \quad (13)$$

Here, I_1 and I_2 are the two invariants of the Green–Lagrange finite strain tensor and C_{ij} are the material coefficients. $G(t)$ in equation (12) is the time-dependent relaxation function. In this work, for a non-integer order time derivative, rather than using time decaying exponentials, which are solution to integer order time derivatives, the Mittag–Leffler function, which is the solution to non-integer order time derivatives was utilized (equation (14)).

$$G(t) = \left\{ 1 - \delta \left[1 - E_\beta \left(-\left(\frac{t}{\tau} \right) \right)^\beta \right] \right\}. \quad (14)$$

Here, δ is the relaxation ratio, $E_\beta(\cdot)$ is the one term Mittag–Leffler function, with β being the order of differentiation with respect to time and τ is the time constant of the model.

The material model obtained contains eight material parameters to be determined, five of them are the instantaneous elastic coefficients, C_{10}, C_{01}, C_{11}, C_{20}, C_{30} which shape the instantaneous relation between second Piola–Kirchoff stress tensor and Green–Lagrange finite strain tensor, and the remaining three are the time dependent coefficients δ, β and τ which, in relaxation form, determine the fading memory of the material.

This material model was implemented to commercial finite element software Msc. Marc-Mentat® 2010 by the user subroutine uelastomer written in FORTRAN. The instantaneous (glassy) strain energy density function was defined in terms of first and second principal stretch ratios, λ_1 and λ_2 as required by the software. The bulk modulus of soft tissue was modeled as 1000 times the initial tangent modulus which turned out to be nearly incompressible material. For relaxation (time dependent) part of the strain energy density function, the numerical value of Mittag–Leffler function was calculated by the approximate formula presented in equation (11). The user subroutine and finite element model using this subroutine were exhaustively checked against known responses and the accuracy was found to be satisfactory [10].

3. Results

In-vivo indentation experiments performed on human fore-arm bulk soft tissues were utilized for determining the capability of non-integer order material model to simulate soft biological tissue material behavior [23], [24]. These experiments are performed by a computer controlled custom-made indenter having a step motor and a loadcell to measure tissue reaction force. Cyclic experiments at 1, 2, 4 and 8 mm/s rates, relaxation and creep experiments for 120 s are performed. Time, indenter tip displacement and tissue reaction force are recorded during each experiment. Although an elliptical tip is used in these experiments to determine in-plane tissue anisotropy, the non-integer order viscoelastic material model presented in this study ignored tissue anisotropy and was isotropic.

Modeling force-relaxation and creep indentation experiments

The non-integer order relaxation and creep functions were tested against experimental force-relaxation and creep data. The Mittag–Leffler function was computed using the exact formula supplied by Podlubny [26]. Optimization toolbox of Matlab® was utilized for Levenberg–Marquardt algorithm to minimize the least square error between the experimental data and model prediction (equation (15)).

$$\mathrm{LSE} = \sqrt{\frac{\sum_{i=1}^{n}(\mathrm{Exp}_i - \mathrm{Model}_i)^2}{\sum_{i=1}^{n}(\mathrm{Exp}_i)^2}} \times 100. \qquad (15)$$

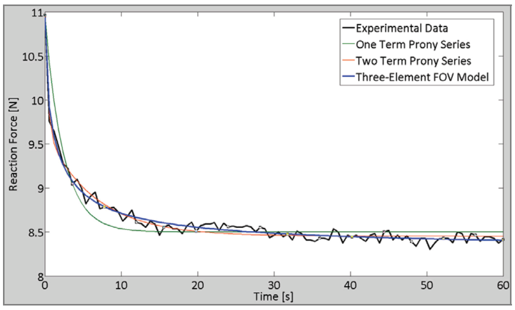
The initial estimates for the material parameters were $\delta = 0.5$, $\eta = 1$ and $\beta = 0.5$.

For comparison purposes the same experimental data was modeled by using the integer order Zener model with one and two exponential decays in the form of Prony series (equation (16) for relaxation and equation (17) for creep with $N = 1$ for one term and $N = 2$ for two terms).

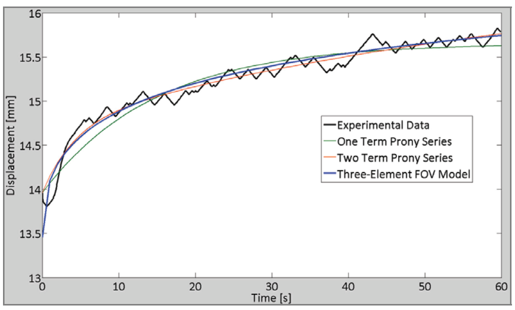
$$G(t) = E_0\left[1 - \sum_{i=1}^{N}\delta_i\left(1 - e^{-\frac{t}{\tau_i}}\right)\right], \qquad (16)$$

$$J(t) = D_0\left[1 + \sum_{i=1}^{N}\delta_i'\left(1 - e^{-\frac{t}{\tau_i'}}\right)\right]. \qquad (17)$$

The experimental data together with integer order one exponential and two exponentials and non-integer order model are presented in Fig. 4. Resulting coefficients and LSE are presented in Tables 1 and 2 for relaxation and creep, respectively.



(a) Relaxation



(b) Creep

Fig. 4. Experimental data modeled with one and two term Prony series and non-integer (fractional) order viscoelastic (FOV) equations a) relaxation, b) creep

Table 1. Parameters identified and resulting LSE for the three models for relaxation

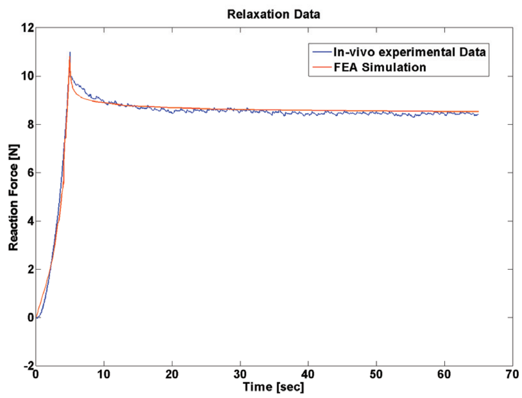
FOV Zener	Value	Unit	Integer order Prony	Two-term Prony	One-term Prony	Unit
E_1	0.591	kN/m	E_0	0.785	0.785	kN/m
E_2	0.192	kN/m	δ_1	0.1194	0.225	–
η	3.44	s	τ_1	0.389	2.39	s
β	0.619	–	δ_2	0.110	–	–
			τ_2	6.49	–	s
LSE	3.44	%	LSE	5.10	8.32	%

Table 2 Parameters identified and resulting LSE for the three models for creep

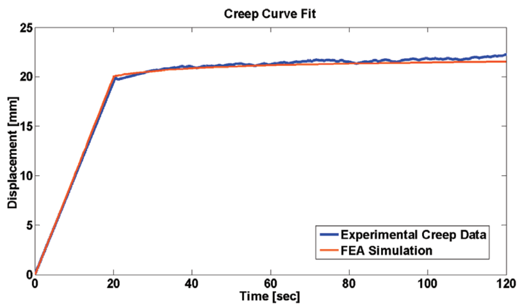
FOV Zener	Value	Unit	Integer order Prony	Two-term Prony	One-term Prony	Unit
E_1	0.548	kN/m	D_0	1.274	1.274	m/kN
E_2	0.193	kN/m	δ_1	0.0552	0.1215	–
η	0.0879	s	τ_1	4.16	14.6465	s
β	0.439	–	δ_2	0.134	–	–
			τ_2	74.1	–	s
LSE	1.95	%	LSE	2.90	3.12	%

Simulation of indentation experiments by finite element model

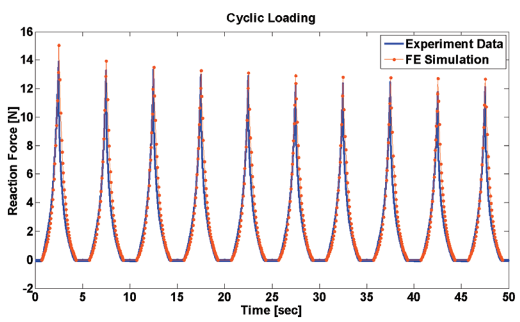
Cyclic loading and unloading, relaxation and creep experimental data was used in inverse finite element modeling with non-integer order viscoelastic material model for the purpose of material parameter identification. The quality of the fit was evaluated using the normalized sum of square error (NSSE) defined in equation (18).



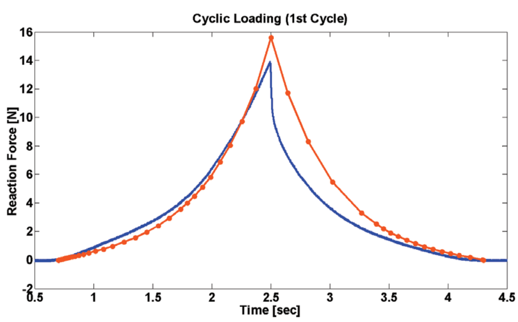
(a) Relaxation



(b) Creep



(c) Cyclic



(d) Cyclic detail

Fig. 5. Experimental data modeled with a single material parameter set extracted from relaxation and creep experiments a) relaxation, b) creep, c) cyclic, d) cyclic detail

$$\mathrm{NSSE} = \sum_{i=1}^{N} \left(\frac{F_{\mathrm{Exp}_i} - F_{FE_i}}{F_{\mathrm{Exp}_{\max}}} \right)^2 . \tag{18}$$

The initial estimates for the nonlinear elastic parameters, C_{ij} were 1 kPa and time dependent parameters were $\tau = 1$ s, $\delta = 0.5$, $\beta = 0.5$. Because of the highly nonlinear nature of the problem a manual search for the material parameters was performed until NSSE reduced to 1 to 2%. The search started with fitting nonlinear elastic parameters based on initial loading data. Then, by using relaxation or creep part the time dependent parameters were determined. Change in time dependent parameters caused initial loading simulations to deviate from experimental data, so they were fine-tuned based on experimental data and finally time dependent parameters were fine-tuned based on changes in nonlinear elastic parameters. This iteration continued a few times till desired NSSE level was reached or in rare cases when NSSE seemed to settle to a value. Then, a single, optimized, parameter set was searched which would simulate cyclic loading, relaxation and creep experiments with desired accuracy, therefore would be a predictive material model for a range of experiments rather than a single curve fit for a single type of experiment. The results of such a parameter set are presented in Fig. 5, simulating relaxation, creep and cyclic loading which was obtained solely from relaxation and creep experiments. All experiments were done on the same volunteer on the same anatomical location at the same date allowing enough time for the tissue to recover. The material parameter set utilized in all simulations is presented in Table 3.

Table 3. Material parameter set and NSSE values for FE simulations

Parameter	Value	Unit
C_{10}	0.1	kPa
C_{01}	0.1	kPa
C_{11}	0.5	kPa
C_{20}	0.5	kPa
C_{30}	0.1	kPa
τ	2	s
δ	0.3	–
β	0.70	–
NSSE (Relax)	1.15	%
NSSE (Creep)	1.15	%
NSSE (Cyclic)	4.5	%

4. Discussion

Viscoelastic models based on non-integer order derivatives are recognized in literature for accurately representing the experimental behavior with less number of parameters and they are reported to have a physical interpretation. In this work, an intermediate mechanical element, the spring-pot that can monotonically and continuously be varied between an elastic solid having perfect memory and a viscous fluid with no memory was reviewed together with non-integer time derivatives. The monotonic variation of spring-pot from elastic solid to viscous fluid, together with its constitutive equation was presented. The dashpot in one of the classical viscoelastic material models, the three element (Zener) model was replaced with a spring-pot. The ability of non-integer order three element model in simulating the force-relaxation and creep indentation experiments performed *in-vivo* on human forearm [23], [24] were compared with integer order viscoelastic models. Integer order model with one Prony series term had three parameters to be identified and it resulted the largest LSE. Integer order model with two Prony series term had five parameters to be identified and LSE was lower compared to one term Prony series model. The non-integer order model had four parameters to be identified and it resulted the lowest LSE. For force-relaxation experiment non-integer order, β, turned out to be 0.619 and for creep experiment 0.439 both being around 0.5 meaning the intermediate element was somewhere in between an elastic solid and a viscous fluid.

The success of one dimensional model simulating the force-relaxation and creep experiments motivated applying the non-integer order models to constitutive equations for soft tissues. A separable nonlinear elastic and non-integer order relaxation in the form of a product was used to relate Green–Lagrange finite strain tensor to second Piola–Kichhoff stress tensor in the commercial finite element program Msc. Marc-Mentat® 2010 by a user subroutine. The material coefficients for the indentation experiments were predicted using relaxation data and optimized for creep and relaxation (i.e., the NSSE for both simulations were equated for a single set of material parameters). This set, when used in cyclic experimental data simulation yielded an NSSE of 4.5% which is acceptable. Although single cycle simulation result does not seem to be very close to the experimental data (Fig. 5d) the ten-cycle simulation (Fig. 5c) was capable of simulating the experimental data accurately, including preconditioning (Mullin's effect). For this specific volunteer and anatomical location the non-integer order resulted to be 0.70 which is closer to a viscous fluid but still having some elastic solid properties.

The finite element simulations with a single set of non-integer order viscoelastic material model parameters approximating three different types of experiments, cyclic loading and unloading, force-relaxation and creep showed that the constitutive equation utilized was not a mere curve-fitting for a single experiment but it has a strong potential for a predictive material model.

There are certain limitations of the current study. The non-integer order model was tested against a very limited number of experimental data. To be able to generalize the results, the proposed material model should be extensively tested. The constitutive equation was a priori assumed to be composed of separable nonlinear hyperelastic and time dependent parts as a product, that is, nonlinear elastic part is independent of time (instantaneous elastic) and time dependent part is independent of strain. Although this assumption is widely used for biological soft tissues as well [6], [13]–[15], [35] its validity should be questioned. The nonlinear elastic part of the constitutive equation was modeled by a special form of a strain energy density function, the James–Green–Simpson material model. This material model although yields good predictions for long chain molecule materials like polymers, rubber and biological soft tissues in finite strain, being a phenomenological material model, it lacks the cause and effect relationship of the observed mechanical behavior. Therefore the material coefficients of the model do not have any physical interpretation in finite strain. Soft biological tissues are known to possess considerable anisotropy as also evident in the experimental data [23], [24] that was used to evaluate the proposed material model. However anisotropy was ignored in proposed material model.

The non-integer order time dependent behavior was found to be a promising candidate for simulating indentation experiment of the soft biological tissues. Structural or micro-structural elastic material models together with non-integer time dependent behavior may supply better insight to observed mechanical behavior of soft biological tissues however obtaining structural or micro-structural information of the soft tissue to be modelled might not be as simple as performing indentation experiments

Acknowledgments

The second author would like to thank Dr. M. Barbara Silver-Thorn (Marquette University) with whom he started the research on soft tissue biomechanics, Prof. Dr. S. Turgut Tümer (Middle East Technical University) who encouraged and supported him to conduct research in biomechanics. The second author dedicates this manuscript to the memory of Dr. H. Cenk Güler, member of the team starting the Biomechanics Laboratory of Mechanical Engineering Department of Middle East Technical University, in which, this research was conducted. This research was partially funded by the Middle East Technical University Scientific Research Funds, grant number BAP-03-02-2010-02.

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