



HAL
open science

On the nonlinear viscoelastic behavior of rubber-like materials: Constitutive description and identification

Adel Tayeb, Makrem Arfaoui, Abdelmalek Zine, Adel Hamdi, Jalel Benabdallah, Mohamed Ichchou

► To cite this version:

Adel Tayeb, Makrem Arfaoui, Abdelmalek Zine, Adel Hamdi, Jalel Benabdallah, et al.. On the nonlinear viscoelastic behavior of rubber-like materials: Constitutive description and identification. International Journal of Mechanical Sciences, 2017, 130, pp.437-447. 10.1016/j.ijmecsci.2017.06.032 . hal-02444061

HAL Id: hal-02444061

<https://hal.science/hal-02444061>

Submitted on 17 Jan 2020

HAL is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.



Distributed under a Creative Commons Attribution 4.0 International License

On the nonlinear viscoelastic behavior of rubber-like materials: Constitutive description and identification

Adel Tayeb^{a,b}, Makrem Arfaoui^a, Abdelmalek Zine^c, Adel Hamdi^a, Jalel Benabdallah^a, Mohamed Ichchou^b

^a*Université de Tunis El Manar, École Nationale d'Ingénieurs de Tunis, LR-11-ES19
Laboratoire de Mécanique Appliquée et Ingénierie, Tunis 1002, Tunisie*

^b*Laboratoire de tribologie et dynamique des systèmes, École Centrale de Lyon, Ecully
69130, France*

^c*Université de Lyon, Institut Camille Jordan, CNRSUMR5208, Département de
Mathématiques et Informatique, École Centrale de Lyon, 36 av. Guy de Collongue,
69134 Ecully Cedex, France*

Abstract

The main concern of this paper is the development of a fully three dimensional viscoelastic model at finite strain to describe nonfactorizable behavior of rubber-like materials within the framework of rational thermodynamics and internal state variable approach such that the second law of thermodynamics in the form of Clausius-Duhem inequality is satisfied. The nonfactorizable aspect of the behavior is introduced via a strain dependent relaxation times. The model is applied to describe the response of the isotropic Pipkin multi-integral viscoelastic model and the BIIR material, several parameters involved are then identified using quasi-static and dynamic experiments thanks to a least-square minimization procedure performed with Matlab software. The proposed model is able to reproduce quasi-static experimental response and show an ability to predict the dynamic response of nonfactorizable rubber-like materials in a wide range of strain.

Keywords: nonfactorizable viscoelasticity, Rubber, Finite-strain, Identification

1. Introduction

It is well known that rubber-like materials exhibit nonlinear viscoelastic behavior over a wide range of strain and strain rates confronted in several

4 engineering applications such as civil engineering, automotive and aerospace
5 industries. This is due to their capacity to undergo high strain and strain
6 rates without exceeding the elastic range of behavior. Further, the time de-
7 pendent properties of these materials, such as shear relaxation modulus and
8 creep compliance, are, in general, functions of the history of the strain or the
9 stress [1]. Therefore, in a wide range of strain, a linear viscoelasticity theory
10 is no longer applicable for such material and new constitutive equations are
11 required to fully depict the behavior of rubber-like materials for quasi-static
12 and dynamic configurations of huge interest in engineering applications.

13 In the literature, several phenomenological models have been developed to
14 describe the nonfactorizable behavior of rubber-like materials, namely the
15 Solid-Liquid viscoelastic model in the series of papers by Chang et al. [2],
16 [3] and Sullivan in [4] for which a generalized measure of deformation has re-
17 placed the strain tensor in the linear Boltzmann convolution integral model
18 and the nonlinear viscoelastic model by Schapery [5] in which the creep com-
19 pliance and the shear relaxation functions were considered stress-dependent
20 and strain-dependent functions respectively and the model of Valanis [6] in
21 which a total thermodynamic formulation led to a constitutive equation de-
22 pending on the deformation via a deformation shift function in analogy with
23 the so-called rheologically simple materials.

24 In the other hand, other models based on the microstructure of the polymeric
25 chain have been proposed such as the model by Knauss et al. [7] in which,
26 following polymer science, time dependent functions were dependent upon
27 the volumetric strain via a strain shift function and the model by Caruthers
28 et al. [8] in which the strain shift function was expressed in terms of the
29 configurational energy of the molecular structure.

30 In this work we shall develop a nonlinear viscoelastic model at finite strain
31 within the framework of rational thermodynamics and the approach of inter-
32 nal state variables, the model is derived through a combination of approaches
33 in [9], [10] and [11] taking into account the dependence of the time depen-
34 dent functions upon the state of the strain. The model's parameters are then
35 identified using data generated from the multi-integral viscoelastic model of
36 Pipkin [12] and experimental data for bromobutyl (BIIR) from [13].

37 This paper is organized as follow: in section 2, a one dimensional nonlinear
38 viscoelastic model is developed using a modified Maxwell rheological model.
39 In the section 3, this model is extended to the fully nonlinear formulation us-
40 ing a nonlinear set of evolution equation of the internal state variables within
41 the rational thermodynamic framework. The shear relaxation modulus is set

42 to be a function upon the invariants of the right Cauchy-Green strain tensor
43 via a strain shift function analogous to the temperature shift function for the
44 thermorheologically simple materials, this choice is motivated experimentally
45 [13] and rheologically [14]. The constitutive equation for the stress is then
46 obtained by resolving the set of nonlinear evolution equations. In section 4,
47 a systematic identification procedure of several parameters involved in the
48 model is highlighted. The optimization problems arising from this identifica-
49 tion procedure are solved by a modified least square minimization algorithm
50 with Matlab software. The section 5 is devoted to the results of this identifi-
51 cation procedure using an experimental characterization of the Bromobutyl
52 BIIR [13] and a theoretical data using the Pipkin model [12]. The capacity
53 of the model to describe the behavior of the material is then outlined.

54 **2. Experimental and rheological motivations**

55 In this **section** we develop the rheological and experimental arguments
56 leading to the proposed finite strain viscoelastic model. To motivate the three
57 dimensional model developed below, we first highlight some experimental
58 results leading to this model and then we consider a suitable modification
59 to the generalized Maxwell rheological model to build the one dimensional
60 nonlinear viscoelastic model.

61 *2.1. Experimental motivation*

62 A significant class of rubbers shows nonfactorizable behavior at low and
63 average range of strain. This phenomenon consists on the dependence of
64 the shear relaxation modulus upon strain level. Several works were dedi-
65 cated to deal with this class of behavior especially the series of papers by
66 [4] and [15]. In a recent work [13], an experimental characterization was
67 carried out with three rubber-like materials: the natural rubber (NR), the
68 Bromobutyl (BIIR) and a mixture of these materials (NR-BIIR). **These** ma-
69 terials showed a dependence of the shear relaxation modulus upon strain.
70 In figure 1 it is plotted the logarithm of the shear relaxation module $G(t)$
71 versus the logarithm of time for two different level of strain 10% and 50% for
72 BIIR material. The shear relaxation modulus shows a dependence upon the
73 strain level which leads according to [16] to a shift in the time with a strain
74 dependent function since the shear relaxation module at any level could be
75 obtained through a combination of a vertical and horizontal translation from
76 the reference curve at a strain level of 10%. Therefore a one dimensional

77 viscoelastic model, taking in consideration these results, is developed in the
 78 next section through a generalization of the Maxwell rheological model.

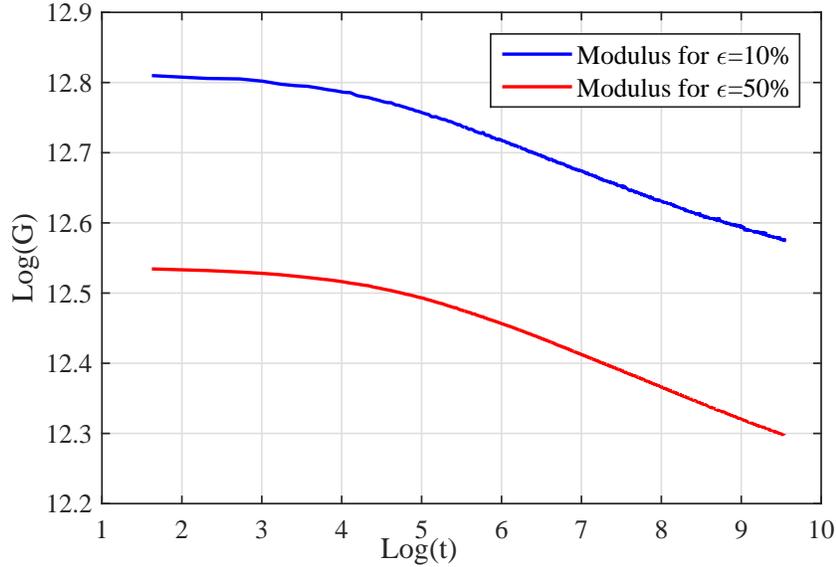


Figure 1: Dependence of the shear relaxation modulus upon strain for BIIR rubber

79 *2.2. Rheological motivation*

80 Before we develop the three-dimensional viscoelastic model, we shall in-
 81 vestigate the following formulation for a standard linear solid of the figure 2.
 82 In this model, σ denotes the total stress, ϵ denotes the total strain, k_i and τ_i
 83 are the parameters of the Maxwell model. Unlike the rheological model used
 84 in [17], the relaxation times τ_i are, due to the experimental result outlined
 85 above, functions of the total strain ϵ . Furthermore, the stress in the spring
 86 of each Maxwell branch is denoted by q_i and its governed by the following
 87 evolution equation.

$$\dot{q}_i + \frac{1}{\tau_i(\epsilon)} q_i = \frac{1}{\tau_i(\epsilon)} k_i \epsilon, \quad q_i|_{t=0} = 0 \quad (1)$$

88 The total stress σ derive directly from the rheological model of figure 2 as
 89 the difference between the elastic equilibrium stress and the non-equilibrium
 90 stresses q_i .

$$\sigma = k_e \epsilon - \sum_i q_i \quad (2)$$

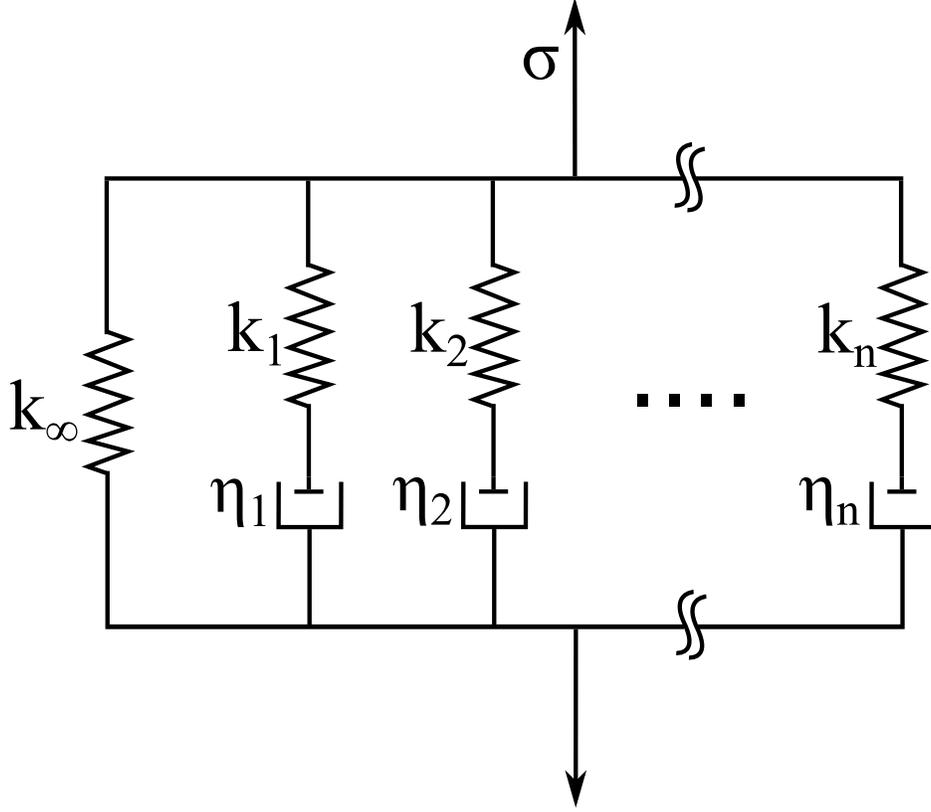


Figure 2: Generalized Maxwell model

91 The time parameters of the Maxwell model are set to be a strain depen-
 92 dent function; this idea follows from the description of thermorheologically
 93 simple materials behavior see [16] and [18], for which all parameters are tem-
 94 perature dependent via a single variable function called *temperature shift-*
 95 *function*. [5] [19] and [15] among others generalized this notion to describe
 96 thermorheologically complex materials behavior where the shift function depen-
 97 dent upon temperature and stress or strain. Hence, the time parameters
 98 take the following form.

$$\tau_i(\varepsilon) = a(\varepsilon) \tau_i \quad (3)$$

99 $a(\varepsilon)$ is a non-negative strain function called strain shift function. Therefore,
 100 the law of evolution of the equation 1 became a linear differential equation
 101 over the reduced time , after considering the form of the time parameters of

102 equation 3.

$$\frac{dq_i}{d\xi} + \frac{1}{\tau_i} q_i = \frac{1}{\tau_i} k_i \varepsilon \quad \text{with} \quad \xi(t) = \int_0^t \frac{dt'}{a(\varepsilon)} \quad (4)$$

103 $\xi(t)$ is an increasing function of time, integration of 4 and substitution of q_i by
 104 its expression into 2 yield the expression of the total stress σ as a Boltzmann
 105 convolution integral of the strain.

106 3. Fully nonlinear viscoelastic model

107 In this section we extend the formulation outlined above to the fully
 108 nonlinear range. Hence, the mechanical framework and the thermodynamic
 109 assumptions leading to the model are outlined. It should be noted, however,
 110 that this model is derived through an isothermal conditions.

111 3.1. Mechanical framework and form of the Helmholtz free energy density

112 Consider a viscoelastic material with reference placement Ω_0 in the ref-
 113 erence configuration \mathbb{C}_0 . It occupies at the time t the placement Ω in
 114 the deformed configuration C_t see figure 3. Let φ denote a macroscopic
 115 motion between the two configurations, which maps any point X in the
 116 reference configuration C_0 to the point x in the deformed configuration.
 117 Let $F(X, t) = \partial x / \partial X$ be the deformation gradient tensor. Likewise, let
 118 $J = \det(F)$ be the jacobian of the deformation gradient tensor. From
 119 the deformation gradient $F(X, t)$ the deformation tensor of Green Lagrange
 120 $E = 1/2(C - I)$, the right and left Cauchy-Green strain tensors $C = F^t F$
 121 and $B = F F^t$ are obtained, together with their principal invariants.

$$I_1 = \text{tr}C, \quad I_2 = \frac{1}{2} [(\text{tr}C)^2 - \text{tr}C^2] \quad \text{and} \quad I_3 = \det(C) = J^2 \quad (5)$$

122 which, otherwise, can be expressed in terms of principal stretches by

$$I_1 = \lambda_1^2 + \lambda_2^2 + \lambda_3^2, \quad I_2 = \lambda_1^2 \lambda_2^2 + \lambda_2^2 \lambda_3^2 + \lambda_1^2 \lambda_3^2 \quad \text{and} \quad I_3 = \lambda_1^2 \lambda_2^2 \lambda_3^2 \quad (6)$$

123 The formulation in the nonlinear range is based on the decomposition of the
 124 gradient $F(X, t)$ into a volume-preserving and pure dilatational part as its
 125 originally proposed by Flory in [20] and used in several works such as [9] and
 126 [21] among others as follow:

$$F = \bar{F} \det(F)^{1/3} I \quad \text{where} \quad \det(\bar{F}) = 1 \quad (7)$$

127 \bar{F} is the volume-preserving gradient tensor, the advantage of such decompo-
 128 sition is its validity near and far away from the thermodynamic equilibrium
 129 [11]. The Cauchy-Green strain tensor associated and the Lagrangian strain
 130 tensor associated with the volume-preserving gradient are expressed as

$$\bar{C} = \bar{F}^t \bar{F} = J^{-2/3} C, \quad \bar{E} = \frac{1}{2} (\bar{C} - I) \quad (8)$$

131 I is the metric tensor in the reference configuration. Furthermore, several
 132 applications of the chain rule lead to the following

$$\frac{\partial \bar{E}}{\partial E} = \frac{\partial \bar{C}}{\partial C} = J^{-2/3} \left[I - \frac{1}{3} C \otimes C^{-1} \right] \quad (9)$$

133 I is the fourth order unit tensor ~~i.e. ($I_{ijkl} = 1$ if $(i = j = k = l)$; else $I_{ijkl} =$~~
 134 ~~0),~~ the sign \otimes designates the tensorial product ~~in the reference basis.~~ Hence,
 135 we postulated an uncoupled free energy density as its expressed in [9] by a
 136 Taylor series in which terms higher than the second order are omitted.

$$\Psi(\bar{C}, Q) = U^0(J) + \bar{\Psi}^0(\bar{C}) - \frac{1}{2} Q : \bar{C} + \Psi_I(Q) \quad (10)$$

137 Q is a second order overstress tensor internal variable akin to the second
 138 Piola-Kirchhoff stress tensor S . The first two terms of the free energy density
 139 of equation 10 are the dilatational and volume-preserving parts of the stored
 140 elastic energy density. The third and fourth terms are responsible for the
 141 time-dependent behavior of the material. Note that $\Psi_I(Q)$ is a convex
 142 function of the internal variable Q . This decomposition of the free energy
 143 density leads to a decomposition in the stress into a deviatoric (shear) and
 144 hydrostatic (bulk) parts.

145 3.2. Rate and constitutive equations

146 The rate equation of the internal variable Q is motivated by the rate
 147 equation 4 of the rheological model in which the elastic stress is replaced by
 148 the deviatoric part of the hyperelastic Second Piola-Kirchhoff stress tensor
 149 as it's expressed in [9] **for which the behavior in bulk is considered purely**
 150 **elastic.**

$$\frac{\partial Q}{\partial \xi} + \frac{1}{\tau} Q = \frac{\gamma}{\tau} DEV \left[2 \frac{\partial \Psi^0(\bar{C})}{\partial \bar{C}} \right] \quad \text{with} \quad \xi(t) = \int_0^t \frac{dt'}{a(\bar{C})} \quad (11)$$

151 As in the previous section, $a(\bar{C})$ is a function of the invariants of the volume-
 152 preserving right Cauchy-Green strain tensor \bar{C} and ξ is referred to as the
 153 reduced time and **its** an **increasing** function of time. The second law of
 154 thermodynamic is expressed in terms of the Clausius-Duhem inequality in
 155 the reference configuration C_0 .

$$-\dot{\Psi} + \frac{1}{2}S : \dot{C} \geq 0 \quad (12)$$

156 Standard arguments [22] and [23] using inequality 12 lead to the expression
 157 of intrinsic dissipation and the second Piola-Kirchhoff stress tensor.

$$-\frac{\partial \Psi(C, Q)}{\partial Q} : \dot{Q} \geq 0 \quad \text{and} \quad S = \frac{1}{2} \frac{\partial \Psi(C, Q)}{\partial C} \quad (13)$$

158 Let ~~$\tau = J\sigma = F S F^t$~~ be the Kirchhoff stress tensor, where σ is the Cauchy
 159 stress tensor and S is the second Piola-Kirchhoff stress tensor. Considering
 160 relations 9, 11 and 13 one could simply lead to the convolution representation
 161 of the second Piola-Kirchhoff stress tensor.

$$S = J^{-2/3} \int_0^\xi g(\xi - \xi') \frac{\partial}{\partial \xi'} DEV \left(2 \frac{\partial \Psi^0(\bar{C})}{\partial \bar{C}} \right) d\xi' + JpC^{-1} \quad (14)$$

162 in which $DEV(\bullet) = (\bullet) - \frac{1}{3}[C : (\bullet)]C^{-1}$ denotes the deviator operator
 163 in the reference configuration. $p = \partial U^0(J)/\partial J$ is the hydrostatic part of
 164 the stress, for an incompressible material, p is an undetermined pressure to
 165 be obtained by the boundary conditions. g is the normalized shear relax-
 166 ation modulus and its a decaying function **of time**, its often expressed by a
 167 power law function or a decaying exponential function. For computational
 168 reasons its more efficient to consider the Cauchy stress tensor rather than
 169 the second Piola-Kirchhoff stress tensor. Application of an integration by
 170 parts to the expression of the second piola-Kirchhoff stress tensor of rela-
 171 tion 14 and considering the relative distortional deformation gradient tensor
 172 $\bar{F}_t(t') = J^{-1/3} \partial \Phi(X, t') / \partial \Phi(X, t)$ the Cauchy stress tensor reads

$$\sigma = \sigma_o^d + \frac{1}{J} dev \int_0^\xi \frac{\partial g(\xi')}{\partial \xi'} (\bar{F}_\xi^{-1}(\xi - \xi') \sigma_o^d(\xi - \xi') \bar{F}_\xi^{-t}(\xi - \xi')) d\xi' + pI \quad (15)$$

173 σ_o designates the elastic instantaneous Cauchy stress tensor and may be
 174 written [23]

$$\sigma_o = \beta_0 I + \beta_1 B + \beta_{-1} B^{-1} \quad (16)$$

175 where $\beta_j = \beta_j(I_1, I_2, I_3)$ are the elastic response functions. In terms of the
 176 free **energy density** they are given by

$$\begin{aligned}\beta_0(I_1, I_2, I_3) &= \frac{2}{J} [I_2\Psi_1^0 + I_3\Psi_3^0] \\ \beta_1(I_1, I_2, I_3) &= \frac{2}{J}\Psi_1^0 \\ \beta_{-1}(I_1, I_2, I_3) &= -2J\Psi_2^0\end{aligned}\quad (17)$$

177 where

$$\Psi_k^0 = \frac{\partial\Psi^0}{\partial I_k}, \quad k = 1, 2, 3 \quad (18)$$

178 The free energy density has an alternative form in terms of the principle
 179 stretches given by

$$\tilde{\Psi}^o(\lambda_1, \lambda_2, \lambda_3) = \Psi^o(I_1, I_2, I_3) \quad (19)$$

180 $\tau_o^d = dev(J\sigma_o)$ is the deviatoric part of the instantaneous elastic Kirchhoff
 181 stress tensor and may be expressed in lights of relations 17 and 18 by

$$\tau_o^d = 2 \left[\frac{1}{3} (I_2\Psi_2^0 - I_1\Psi_1^0) I + \Psi_1^0 B - I_3\Psi_2^0 B^{-1} \right] \quad (20)$$

182 Or in lights of relation 19 in the base of principle stretches by

$$\tau_{oi}^d = \lambda_i \tilde{\Psi}_i^o - \frac{1}{3} \sum_{j=1}^3 \lambda_j \tilde{\Psi}_j^o \quad (21)$$

183 where $\tilde{\Psi}_i^o$ refers to the derivative $\tilde{\Psi}^o$ with respect to λ_i . The first term of
 184 the right hand side of 15 designates the instantaneous elastic response of the
 185 material, the second one denotes the time dependent part of the material
 186 whereas the third one is the hydrostatic pressure. For an incompressible ma-
 187 terial relation 15 holds with $J = 1$ and $\bar{F}_t(t') = F_t(t') = \partial\Phi(X, t')/\partial\Phi(X, t)$.
 188 Henceforth, the material is considered incompressible so that the Cauchy
 189 stress tensor σ and the Kirchhoff stress tensor τ are the same quantity.

190 4. Model identification

191 In this section, a systematic identification of the material parameters for
 192 an incompressible nonfactorizable viscoelastic constitutive equation at finite
 193 strain is highlighted. This procedure relies on the separate identification
 194 of hyperelastic potential, viscoelastic kernel and the reduced time function.

195 Considering the form of the constitutive equation 14, each characteristic
 196 function identification reduces to the solution of a nonlinear optimization
 197 problem. The identification procedure is carried out considering homogenous
 198 (in space) uniaxial transformations of simple extension and pure shear. For
 199 such transformation, assuming incompressibility, in the material basis the
 200 deformation gradient tensor may be specified as

$$F(t) = \text{diag}(\lambda_1(t), \lambda_2(t), \lambda_3(t)) \quad (22)$$

201 The condition of incompressibility $J = 1$ leads to the following expression of
 202 the deformation gradient tensor

$$F(t) = \text{diag}(\lambda(t), \lambda^{-1/2}(t), \lambda^{-1/2}(t)) \quad (23)$$

203 For simple extension and

$$F(t) = \text{diag}(\lambda(t), \lambda^{-1}(t), 1) \quad (24)$$

204 For pure shear. In both cases only one component of the stress remains and
 205 the indeterminate hydrostatic pressure is eliminated i.e.

$$\sigma_2 = \sigma_3 = 0 \quad \text{and} \quad \sigma_1 = \sigma_1 - \sigma_2 \quad (25)$$

206 From 15 and 25 the expression of the stress follows for these histories of
 207 deformations considered.

$$\sigma = \sigma_{01}^d - \sigma_{02}^d + \text{dev} \int_0^\xi \frac{\partial g(\xi')}{\partial \xi'} \left(\left[\frac{\lambda^2(\xi)}{\lambda^2(\xi - \xi')} \sigma_{01}^d \right] - \left[\frac{\lambda(\xi - \xi')}{\lambda(\xi)} \sigma_{02}^d \right] \right) d\xi' \quad (26)$$

208 For the simple extension and

$$\sigma = \sigma_{01}^d - \sigma_{02}^d + \text{dev} \int_0^\xi \frac{\partial g(\xi')}{\partial \xi'} \left(\left[\frac{\lambda^2(\xi)}{\lambda^2(\xi - \xi')} \sigma_{01}^d \right] - \left[\frac{\lambda^2(\xi - \xi')}{\lambda^2(\xi)} \sigma_{02}^d \right] \right) d\xi' \quad (27)$$

209 For pure shear. The first two terms of relations 26 and 27 refer to the principle
 210 components of the deviatoric instantaneous elastic part of the stress which
 211 can be obtained from the equilibrium deviatoric elastic stress via:

$$\sigma_{0i}^d = \frac{G_0}{G_\infty} \sigma_{\infty i}^d \quad (28)$$

212 in which G_0 and G_∞ refer to the instantaneous and equilibrium shear re-
 213 laxation modulus see [24] and [25], whereas the integral term depicts the
 214 dissipative or the time dependent part of the stress. A general identifica-
 215 tion procedure could be applied separately to each component of the stress.
 216 Hence, let $\Lambda = [\Lambda_1, \Lambda_2, \dots, \Lambda_m]^t$ be the vector of experimental input data
 217 and $\Theta = [\Theta_1, \Theta_2, \dots, \Theta_m]^t$ be the vector of corresponding experimental re-
 218 sponse. For each component of the stress the response function is written
 219 $F(\Lambda, p) : \mathbb{R} \times \mathbb{R}^n \rightarrow \mathbb{R}$ in which $p = [p_1, p_2, \dots, p_n]^t$ is a vector of material
 220 parameters. The objective function is defined by the squared 2-norm

$$S_F(p) : \|F(\Lambda, p) - \Theta\|_2^2 = \sum_{i=1}^m [F(\Lambda_i, p) - \Theta_i]^2 \quad (29)$$

221 The identification procedure turns out into a minimization problem which
 222 reads as follow

$$\min_p S_F(p) \quad (30)$$

223 4.1. Identification of the hyperelastic potential

224 The free energy density Ψ^o is a function of, either, the invariants of the
 225 right Cauchy-Green strain tensor or the principles stretches. The condition
 226 of incompressibility reads

$$\lambda_1 \lambda_2 \lambda_3 = 1 \quad \text{or} \quad I_3 = J^2 = 1 \quad (31)$$

227 The general form of Mooney and Rivlin [26] free energy density is considered
 228 which reads for an incompressible hyperelastic material as follow

$$\Psi^o(I_1, I_2) = \sum_{i,j} c_{ij} (I_1 - 3)^i (I_2 - 3)^j \quad (32)$$

229 c_{ij} are the material parameters of the free **energy density** which usually should
 230 satisfy the stability conditions which ensure an admissible response of the
 231 model for any process see [27] these conditions are expressed as follow:

$$\frac{\partial \Psi^o}{\partial I_1} > 0 \quad \text{and} \quad \frac{\partial \Psi^o}{\partial I_2} \geq 0 \quad (33)$$

232 In the case of uniaxial experiment, the nominal stress which is the measured
 233 quantity, actual force over reference area, and the principle stretch are related

234 through the free energy Ψ^o by the equation

$$\mathbf{\Pi}^o = \frac{\partial \Psi^o}{\partial \lambda} = \sum_{i,j} c_{ij} \phi(i, j, \lambda) \quad (34)$$

235 where $\phi(i, j, \lambda)$ is a nonlinear function of i, j and λ

$$\phi(i, j, \lambda) = 2i \left(\lambda - \frac{1}{\lambda^2} \right) \left(\lambda^2 + \frac{2}{\lambda} - 3 \right)^{i-1} \left(2\lambda + \frac{1}{\lambda^2} - 3 \right)^j + 2j \left(1 - \frac{1}{\lambda^3} \right) \left(\lambda^2 + \frac{2}{\lambda} - 3 \right)^i \left(2\lambda + \frac{1}{\lambda^2} - 3 \right)^{j-1} \quad (35)$$

236 For the simple extension and

$$\phi(i, j, \lambda) = 2(i+j) \left(\lambda - \frac{1}{\lambda^3} \right) \left(\lambda^2 + \frac{1}{\lambda^2} - 2 \right)^{i+j-1} \quad (36)$$

237 For pure shear. An alternative useful representation of equation 34 with re-
238 spect to the identification procedure is used.

239 Let $c^t = \{c_{01}, \dots, c_{0j}, c_{10}, \dots, c_{1j}, \dots, c_{i0}, \dots, c_{ij}\}$ be the vector of material pa-
240 rameters, Φ be a matrix representation of the function $\phi(i, j, \lambda)$ and $\mathbf{\Pi}^e$ the
241 discrete vector of nominal stress. Equation 34 became

$$\mathbf{\Pi}^e = \Phi c \quad (37)$$

242 The identification of the material parameters c_{ij} is performed using data for
243 simple extension and pure shear simultaneously. Therefore, a modification of
244 the objective function 29 is adopted [28]. The new objective function reads
245 as follow

$$\min_{c \in \mathbb{R}^{ij}} \left(\left\| \Phi^{se} c - \tilde{\mathbf{\Pi}}^{se} \right\|_2^2 + \left\| \Phi^{ps} c - \tilde{\mathbf{\Pi}}^{ps} \right\|_2^2 \right) \quad (38)$$

246 The superscript *se* and *ps* refers to the simple extension and pure shear
247 respectively. $\tilde{\mathbf{\Pi}}$ denotes the recorded experimental nominal stress vector. A
248 least square minimization procedure is then employed under conditions 33
249 using Matlab software to reach the numerical values of c_{ij} . The results of
250 this identification and its efficiency are discussed in the latter section of this
251 work.

252 4.2. Identification of the viscoelastic kernel

253 The time dependent part of the stress is characterized by the shear relax-
254 ation function $G(\xi)$ which is a decaying positive function of the reduced time

255 ξ . Its often expressed by, either, a sum of decaying exponential functions
 256 called Prony series function or a power law functions. This identification
 257 is performed using experimental results from relaxation tests and dynamic
 258 tests in the linear range of behavior so that the reduced time is equal to the
 259 real time $\xi = t$ and the behavior of the material is described by the single
 260 convolution integral of Boltzmann:

$$\sigma(t) = \int_0^t G(t-t') \dot{\epsilon}(t') dt' \quad (39)$$

261 ϵ is the linearized strain tensor.

262 4.2.1. Identification from relaxation test

263 The relaxation test is performed in shear deformation. The strain is
 264 suddenly increased to a value ϵ_o and kept constant

$$\epsilon(t) = H(t) \epsilon_o \quad \text{with} \quad H(t) = \begin{cases} 0, & t < 0 \\ 1, & t > 0 \end{cases} \quad (40)$$

265 From equations 39 and 40 the shear relaxation modulus follows

$$G(t) = \frac{\sigma(t)}{\epsilon_o} \quad (41)$$

266 In this work we adopted the Prony series form of the shear relaxation modulus

$$G(t) = G_\infty + \sum_{i=1}^N G_i \exp\left(-\frac{t}{\tau_i}\right) \quad (42)$$

267 G_∞ denotes the long term shear relaxation modulus, G_i $i = 1, \dots, N$ are the
 268 coefficients of the Prony series and τ_i $i = 1, \dots, N$ are the relaxation time
 269 constants. Furthermore, in order to avoid the ill-conditioning of the opti-
 270 mization problem the set of the relaxation times τ_i are a-priori fixed as one
 271 time constant per decade in the logarithmic time scale for the experimen-
 272 tal time window [29] and [30]. The optimization problem arising from the
 273 identification of the N -terms Prony coefficients is

$$\min_{\{G\} \in \mathbb{R}^N} \left\| \Gamma \{G\} - \hat{G} \right\|_2^2 \quad (43)$$

274 where $\Gamma \in \mathbb{R}^{MN}$ is the matrix representation of relation 42 taking the fol-
 275 lowing form

$$\Gamma = \begin{bmatrix} 1 & \exp(-t_1/\tau_1) & \dots & \exp(-t_1/\tau_N) \\ 1 & \exp(-t_2/\tau_1) & \dots & \exp(-t_2/\tau_N) \\ \dots & \dots & \dots & \dots \\ 1 & \exp(-t_M/\tau_1) & \dots & \exp(-t_M/\tau_N) \end{bmatrix} \quad (44)$$

276 $t = \{t_1, \dots, t_M\}$ are the discrete time instants and $\hat{G} = \{\hat{G}_1, \dots, \hat{G}_M\}$ are
 277 the corresponding experimental values of the shear relaxation modulus using
 278 relation 41. A linear least square algorithm is used to solve the optimization
 279 problem 43 using Matlab software.

280 4.2.2. Identification from dynamic tests

281 The dynamic tests are performed using a cylindrical shear sheet loaded by
 282 a sinusoidal deformation without a predeformation and with small amplitude

$$\varepsilon(t) = \varepsilon_o \exp(j\omega t) \quad \text{with } \varepsilon_o \ll 1 \quad (45)$$

283 ω is the circular frequency and j is the unit imaginary number. Hence, from
 284 equations 39 and 45 the stress-strain relation follows

$$\sigma = G^* \varepsilon_o \quad (46)$$

285 G^* is the complex dynamic shear modulus, its real and imaginary parts are
 286 denoted G' and G'' are called storage and loss modulus respectively and may
 287 be obtained by a Fourier transform of equation 42 and given by :

$$\begin{aligned} G' &= G_\infty + \sum_{i=1}^N G_i \frac{(\tau_i \omega)^2}{1+(\tau_i \omega)^2} \\ G'' &= \sum_{i=1}^N G_i \frac{\tau_i \omega}{1+(\tau_i \omega)^2} \end{aligned} \quad (47)$$

288 As mentioned in the previous section, the relaxation times τ_i are a-priori fixed
 289 as one time constant per decade in the logarithmic scale of time. Thereby,
 290 both storage and loss modulus are linear with respect to the N -terms Prony
 291 coefficients. The arising optimization problem from this identification proce-
 292 dure reads

$$\min_{\{G\} \in \mathbb{R}^N} \left(\left\| \Gamma' \{G\} - \hat{G}' - G_\infty \right\|_2^2 + \left\| \Gamma'' \{G\} - \hat{G}'' \right\|_2^2 \right) \quad (48)$$

293 G_∞ is directly identified from the storage modulus curve as $\omega \rightarrow 0$. \hat{G}' and
 294 \hat{G}'' are the experimental vectors of storage and loss modulus, as recorded
 295 by the DMA machine, respectively. Γ' and Γ'' are two M by N matrices
 296 representing equation 47 and can be expressed through the relaxation time
 297 constants and the discrete frequency vector by

$$\Gamma' = \begin{bmatrix} \frac{(\tau_1\omega_1)^2}{1+(\tau_1\omega_1)^2} & \cdots & \frac{(\tau_N\omega_1)^2}{1+(\tau_N\omega_1)^2} \\ \cdots & \cdots & \cdots \\ \frac{(\tau_1\omega_M)^2}{1+(\tau_1\omega_M)^2} & \cdots & \frac{(\tau_N\omega_M)^2}{1+(\tau_N\omega_M)^2} \end{bmatrix}$$

$$\Gamma'' = \begin{bmatrix} \frac{\tau_1\omega_1}{1+(\tau_1\omega_1)^2} & \cdots & \frac{\tau_N\omega_1}{1+(\tau_N\omega_1)^2} \\ \cdots & \cdots & \cdots \\ \frac{\tau_1\omega_M}{1+(\tau_1\omega_M)^2} & \cdots & \frac{\tau_N\omega_M}{1+(\tau_N\omega_M)^2} \end{bmatrix} \quad (49)$$

298 The optimization problem of equation 48 is an ill-posed problem [31]. There-
 299 fore, a Tikhonov [32] regularization method was employed to solve this sys-
 300 tem. The results of this identification using randomly perturbed simulated
 301 and real experimental data are shown in the latter section of this paper.

302 4.3. Identification of the reduced time function

303 Once the hyperelastic potential and the viscoelastic kernel are identified,
 304 the problem of determining the reduced time function can be addressed.
 305 This identification relies on the discretization of the stress-strain relation
 306 (equation 15) with respect to the time. let $t = \{t_1, \dots, t_M\}$ be the discrete
 307 experimental time vector and $\xi = \{\xi_1, \dots, \xi_M\}$ be the corresponding reduced
 308 time vector, Δt is the experimental time increment and $\Delta \xi$ is the reduced
 309 time increment. The general form of this discretization formula [33] for a
 310 nonlinear viscoelastic behavior as follow is described in equation 50. The
 311 identification of the reduced time vector ξ is performed thanks to a recursive
 312 dichotomy algorithm applied to the error between the discretized stress 50
 313 and the experimental stress $\tilde{\sigma} = \{\tilde{\sigma}_1, \dots, \tilde{\sigma}_M\}$.

$$\sigma(t_{n+1}) = \sigma_o^d(t_{n+1}) - \sum_{i=1}^N \sigma_i^d(t_{n+1}) + pI$$

$$\sigma_i^d(t_n) = \frac{g_i}{\tau_i} \int_0^\xi \bar{F}_\xi^{-1}(\xi - \xi') \sigma_o^d(\xi - \xi') \bar{F}_\xi^{-t}(\xi - \xi') \exp\left(-\frac{\xi'}{\tau_i}\right) d\xi'$$

$$\sigma_i^d(t_{n+1}) = \alpha_i g_i \sigma_o^d(t_{n+1}) + \beta_i g_i \hat{\sigma}_o^d(t_n) + \gamma_i \hat{\sigma}_i^d(t_n) \quad (50)$$

with

$$\gamma_i = \exp\left(-\frac{\Delta \xi}{\tau_i}\right); \alpha_i = 1 - \frac{\tau_i}{\Delta \xi} (1 - \gamma_i); \beta_i = \frac{\tau_i}{\Delta \xi} (1 - \gamma_i) - \gamma_i$$

$$\hat{\sigma}_j^d(t) = \bar{F}_t^t(t + \Delta t) \sigma_j^d(t) \bar{F}_t^t(t + \Delta t); j = 0, 1..N$$

314 Once the reduced time vector ξ is obtained the identification of the reduced
 315 time function $a(C)$ can be addressed since it is the inverse of the derivative
 316 of the reduced time with respect to real time:

$$\frac{1}{a} = \frac{d\xi}{dt} \quad (51)$$

317 The derivative in equation 51 is obtained numerically since the reduced time
 318 and the real time are two discrete vectors. Hence, one leads to the numerical
 319 vector of function $a(C) : a = \{a_1, \dots, a_M\}$. Furthermore, a sufficient condi-
 320 tion on this function with respect to the second principle of thermodynamics
 321 in terms of Clausius-Duhem inequality is to adopt a positive function of the
 322 invariants of the right Cauchy-Green strain tensor [14]. A suitable form of
 323 this function is denoted by:

$$a(C) = \exp[-c_1(I_1 - 3) - c_2(I_2 - 3)] \quad (52)$$

324 where c_1 and c_2 are two positive material parameters to be fitted using a
 325 nonlinear curve fitting algorithm with Matlab software.

326 5. Identification of the model using data from the Pipkin isotropic 327 model

328 In this section, the capacity of the proposed model to depict the response
 329 of other complicated viscoelastic models is presented. The main concern is
 330 to reformulate a complicated model namely the isotropic viscoelastic model
 331 by Pipkin [12] in the form of our simple model presented herein. To this end
 332 the identification procedure outlined above is applied using data generated
 333 from the isotropic viscoelastic model proposed by Pipkin [12] see equations
 334 53 and 54. Data were generated from the stress-strain relation in the case of
 335 simple extension and pure shear experiments. Several strain histories were
 336 considered to provide a complete description of the behavior. The hyperelas-
 337 tic potential was identified using data of simple extension and pure shear at
 338 equilibrium, the relaxation function was obtained using a relaxation test per-
 339 formed in simple extension and the reduced time was calculated using mono-
 340 tonic test in simple extension for different strain rates. The identification
 341 procedure is validated by predicting the behavior in pure shear monotonic
 342 tests for different strain rates.

343 *5.1. Pipkin isotropic model*

344 Pipkin [12] proposed a third order development of the tensorial response
 345 function Q for an isotropic incompressible material. The principle of material
 346 indifference requires that the Cauchy stress tensor takes the following form:

$$\sigma = RQR^t + pI \quad (53)$$

347 R is the rotation tensor obtained from the polar decomposition of the trans-
 348 formation gradient tensor F and p is the indeterminate parameter due to
 349 incompressibility. The third functional development of Q reads

$$\begin{aligned} Q(t) = & \int_0^t r_1(t-t') \dot{E}(t') dt' + \int_0^t \int_0^t r_2(t-t', t-t'') \dot{E}(t') \dot{E}(t'') dt' dt'' + \\ & \int_0^t \int_0^t \int_0^t r_3(t-t', t-t'', t-t''') \text{tr} \left[\dot{E}(t') \dot{E}(t'') \right] \dot{E}(t''') dt' dt'' dt''' + \\ & \int_0^t \int_0^t \int_0^t r_4(t-t', t-t'', t-t''') \dot{E}(t') \dot{E}(t'') \dot{E}(t''') dt' dt'' dt''' \end{aligned} \quad (54)$$

350 r_i $i = 1..4$ are the relaxation kernels expressed by a decaying exponential
 351 functions and $\dot{E}(t)$ is the time derivative of the Green-Lagrange deformation
 352 tensor $E = 1/2(C - I)$. Expression of r_i according to [34] is reported in
 353 equation 55, the choice of $r_2(t_1, t_2) = 0$ is motivated by thermodynamic
 354 arguments to ensure the positivity of the free energy density. Further argu-
 355 ments could be found in [34] and references therein.

$$\begin{cases} r_1(t) = a_1 + b_1 \exp(c_1 t) \\ r_2(t_1, t_2) = 0 \\ r_3(t_1, t_2, t_3) = a_3 + b_3 \exp(c_3(t_1 + t_2 + t_3)) \\ r_4(t_1, t_2, t_3) = b_4 \exp(c_4(t_1 + t_2 + t_3)) \end{cases} \quad (55)$$

356 A crucial choice of the parameters a_i , b_i and c_i enables us to describe the
 357 behaviour of the material for any given strain history.

358 *5.2. Identification results*

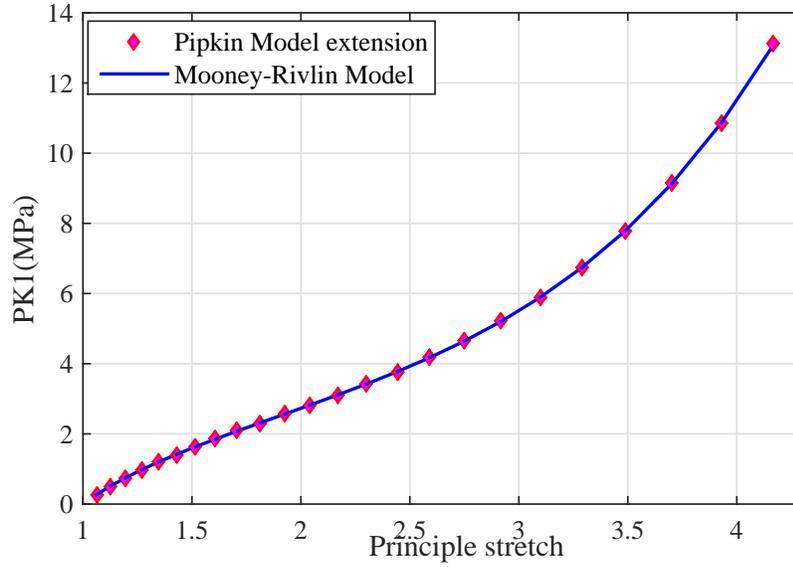
359 *5.2.1. Hyperelastic potential*

360 The identification of the free energy density requires data at equilibrium
 361 in the case of simple extension and pure shear experiments. Hence, data were
 362 generated by omitting the time-dependent part of the stress. Considering the
 363 incompressibility of the behaviour of equations 23 and 24 its straightforward
 364 to obtain from 54 the relations for the equilibrium stress

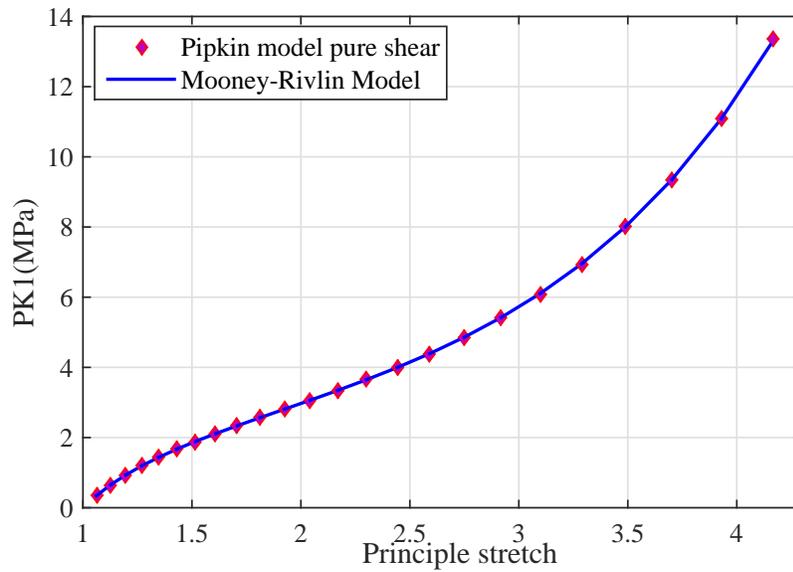
$$\sigma = \left(\lambda^2 - \frac{1}{\lambda} \right) \left[\frac{a_1}{2} + \frac{a_3}{8} \left(\lambda^4 - 2\lambda^2 - \frac{4}{\lambda} + \frac{2}{\lambda^2} + 3 \right) \right] \quad (56)$$

³⁶⁵ in the case of simple extension and

$$\sigma = \left(\lambda^2 - \frac{1}{\lambda^2} \right) \left[\frac{a_1}{2} + \frac{a_3}{8} \left(\lambda^4 - 2\lambda^2 - \frac{2}{\lambda^2} + \frac{1}{\lambda^4} + 2 \right) \right] \quad (57)$$



(a) Equilibrium stress for simple extension



(b) Equilibrium stress for pure shear

Figure 3: Equilibrium stresses versus principle stretch for the Pipkin model (diamond) and the Mooney-Rivlin model (solid curve)

g_i	$\tau_i(s)$
$6.25 \cdot 10^{-2}$	2.003
$2.84 \cdot 10^{-5}$	14.06
$1.12 \cdot 10^{-4}$	82.76

Table 1: Prony series parameters

366 For the simple shear. Results of the identification using the generalized
367 Mooney-Rivlin free energy density in terms of the first Piola-Kirchhoff stress
368 are reported in figure 3 for simple extension and pure shear experiments. A
369 second order generalized Mooney-Rivlin potential, in relation 32, was satis-
370 factory to describe the hyperelastic part of the Pipkin model.

371 *5.2.2. Viscoelastic kernel*

372 The identification of the Prony series requires shear relaxation data at
373 low level of strain. To this end, a Heaviside strain history of relation 40
374 is considered. Introduction of this strain history into 53 and 54 yields the
375 relaxation stress-strain relationship.

$$\sigma(t) = \frac{r_1(t)}{2} \left(\lambda^2 - \frac{1}{\lambda^2} \right) + \frac{r_3(3t)}{8} \left(\lambda^2 - \frac{1}{\lambda^2} \right) \left(\lambda^4 - 2\lambda^2 - \frac{2}{\lambda^2} + \frac{1}{\lambda^4} + 2 \right) + \frac{r_4(3t)}{8} \left(\lambda^2 - \frac{1}{\lambda^2} \right) \left(\lambda^4 - 3\lambda^2 - \frac{3}{\lambda^2} + \frac{1}{\lambda^4} + 4 \right) \quad (58)$$

376 In figure 4 are reported curves of the normalized shear relaxation modulus
377 versus time for four different levels of strain. Its well shown that the hypoth-
378 esis of separability doesnt hold for the Pipkin model since the normalized
379 shear relaxation modulus depends upon strain level. It should be noted,
380 however, that the identification procedure was performed using results of the
381 5% level of strain. Prony series parameters are reported in Table 1.

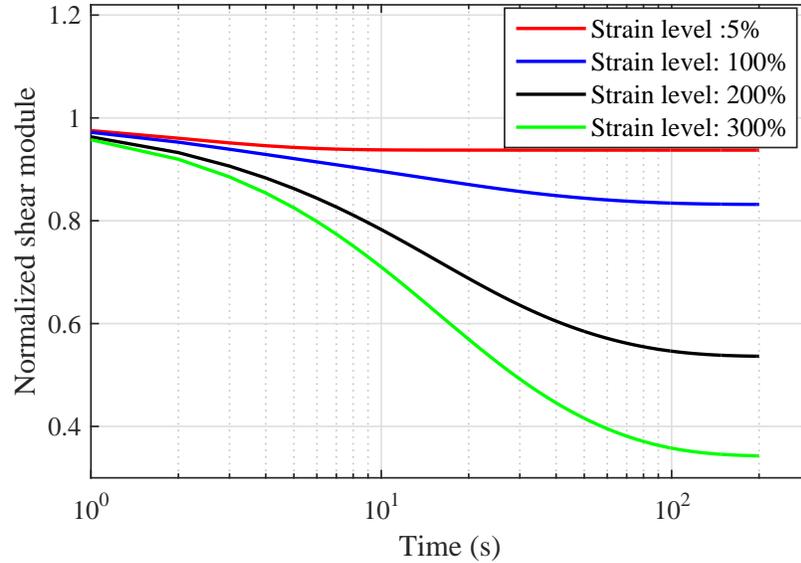


Figure 4: Normalized shear relaxation modulus of Pipkin model versus time for four different strain levels.

382 *5.2.3. Reduced time function*

383 In this part monotonic tests of simple extension and pure shear were
 384 generated from the Pipkin model. Simple extension test was used in the
 385 identification of the reduced time function whereas pure shear test was used
 386 in the validation of the results. For computational convenience with respect
 387 to the multi-integral form involved in 54, the principle stretch corresponding
 388 to a monotonic test was set to be an increasing exponential function of time
 389 of the form:

$$\lambda(t) = \exp(\alpha t) \quad (59)$$

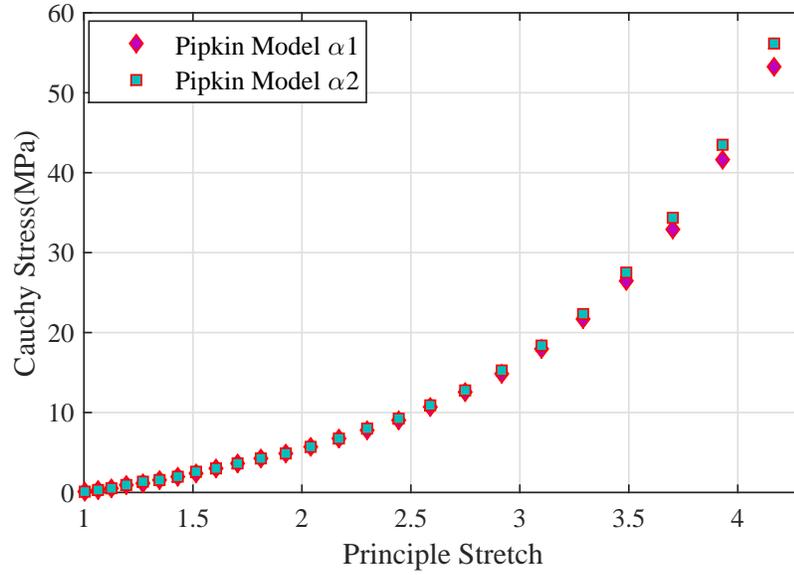
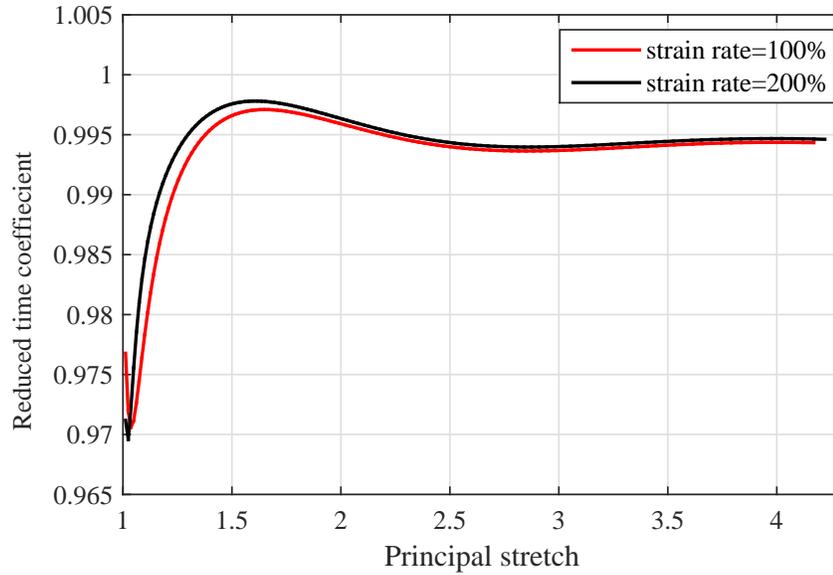
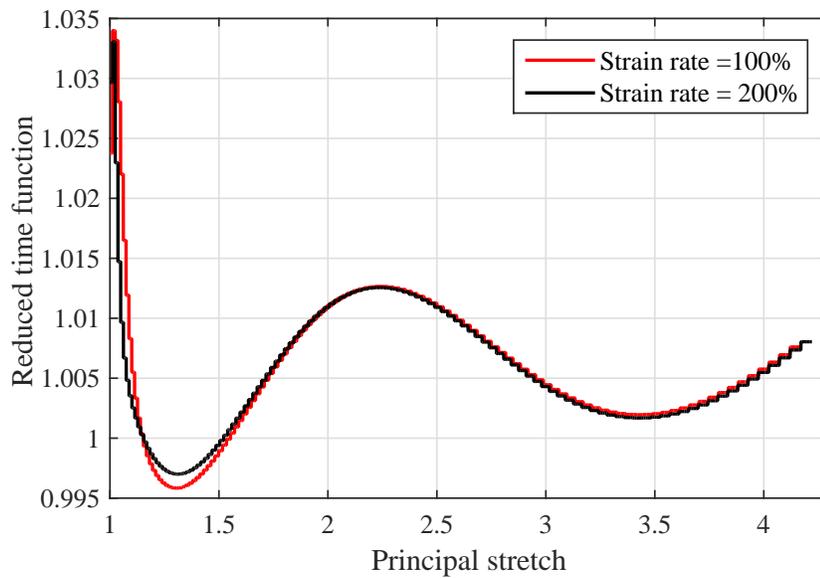


Figure 5: Simple extension Cauchy stress versus principle stretch for two different strain rates $\alpha_1 = 1.19 \cdot 10^{-2}$ and $\alpha_2 = 6 \cdot 10^{-3}$



(a) Reduced time ratio versus principle stretch



(b) Reduced time function versus principle stretch

Figure 6: reduced time function and reduced time ratio versus principle stretch for two strain rates $\alpha_1 = 1.19 \cdot 10^{-2}$ and $\alpha_2 = 6 \cdot 10^{-3}$

390 where α is a positive constant could be interpreted as a strain rate. Re-
 391 placing the principle stretch in 54 by its expression yields the expression of
 392 the Cauchy stress. For each test, two different strain rates were considered
 393 $\alpha_1 = 1.19 \cdot 10^{-2}$ and $\alpha_2 = 6 \cdot 10^{-3}$. Data for simple extension Cauchy stress
 394 are plotted versus principle stretch in figure 5. Hence, the identification pro-
 395 cedure highlighted above was applied to identify the reduced time function.
 396 Results are reported in figure 6 by means of the reduced time ratio $\xi(t)/t$
 397 and the reduced time function $a(C)$ for the two strain rates considered. The
 398 reduced time function is obtained numerically via a numerical derivation of
 399 the reduced time with respect to time. It shows a significant dependence
 400 upon strain level which is consistent with the results shown in Figure 4. Fur-
 401 thermore, this function is independent of the strain rate which motivate the
 402 choice of the form of the reduced time function of equation 52. The capacity
 403 of the nonlinear viscoelastic model developed herein is evaluated by predict-
 404 ing the behaviour of the Pipkin model using the parameters identified in this
 405 section. In order to avoid a division by small value of the force when the
 406 principle stretch is near to one, a modified relative error formula was used as
 407 proposed in [35]

$$err_i = \frac{|\sigma_i - \sigma_i^P|}{\max\{0.5, \sigma_i^P\}} \quad (60)$$

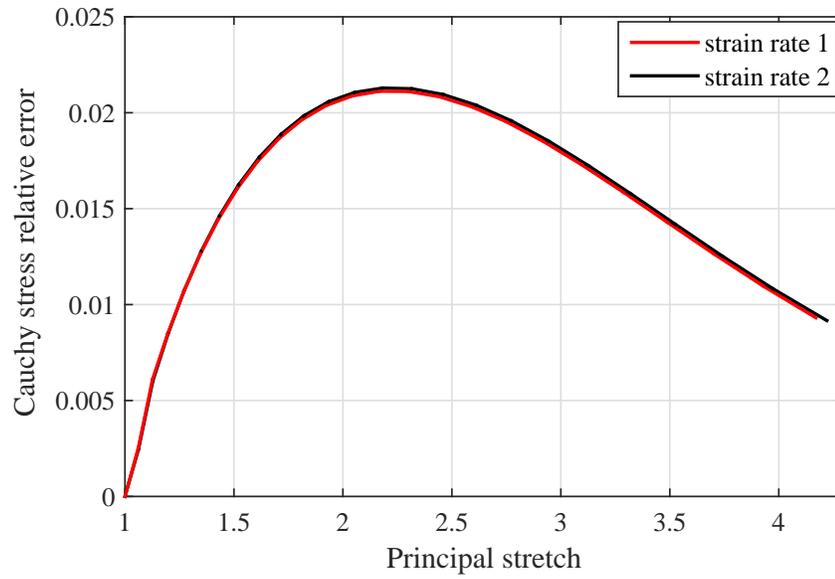


Figure 7: Relative error of the predicted Cauchy stress of the Pipkin model for pure shear experiment

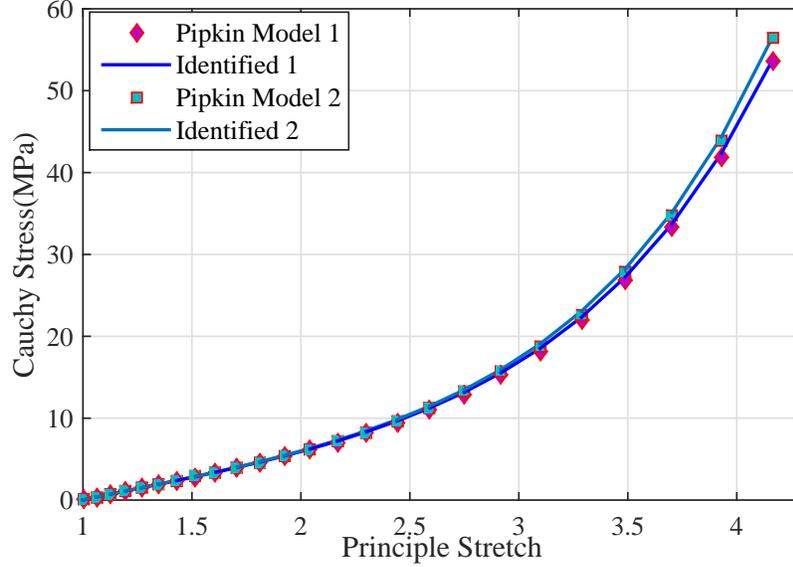
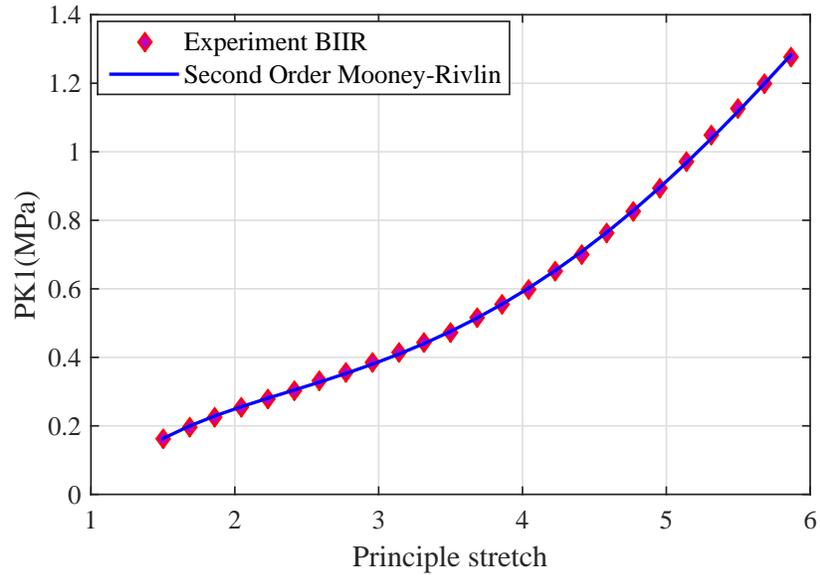


Figure 8: Pure shear Cauchy stress for the model (solid curve) and the Pipkin model (diamond and square)

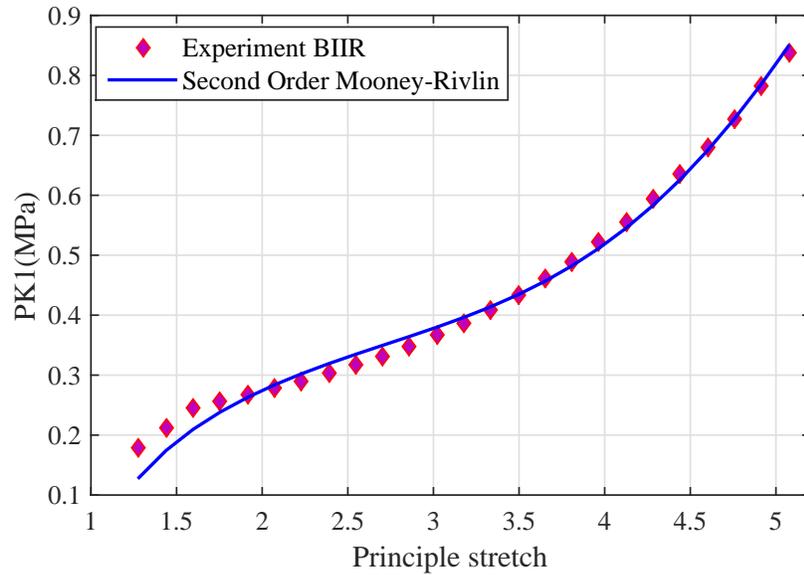
408 in which σ_i is the Cauchy stress computed using 15 and σ_i^P is the Pipkin
 409 Cauchy stress computed using 53 and 54. This function is plotted versus the
 410 principle stretch in figure 7 in the case of pure shear experiment. For the
 411 two strain rates considered, the relative error remains under . In figure 8 is
 412 plotted the Cauchy stress versus principle stretch for the Pipkin model and
 413 the proposed model.

414 6. Application of the identification procedure to experimental data

415 In this section, the identification procedure outlined in section 4 is used to
 416 identify the parameters of the proposed model using experimental data for a
 417 bromobutyl (BIIR) rubber material. Experimental data used here are those
 418 from [13], in which a complete experimental characterization was performed
 419 to obtain the response of the material for several strain history configuration
 420 and several temperatures. In what follows, results of the identification of the
 421 model' parameters are highlighted and discussed.



(a) Equilibrium stress for simple extension



(b) Equilibrium stress for pure shear

Figure 9: Equilibrium stresses versus principle stretch: Experimental (diamond) and the identified Mooney-Rivlin model (solid curve)

g_i	$\tau_i(s)$
$4.46 \cdot 10^{-3}$	14.79
$3.77 \cdot 10^{-2}$	125.71
$5.69 \cdot 10^{-2}$	460.7
$5.84 \cdot 10^{-2}$	1761.6
$8.76 \cdot 10^{-2}$	9598.5

Table 2: Prony series parameters for BIIR rubber

423 The identification of the free energy density coefficients of relation 38 is
424 performed under stability conditions of the relation 33 using Matlab soft-
425 ware. A second order Mooney-Rivlin potential was found to be satisfactory
426 with respect to the value of the total relative error of the stress for simple
427 extension and pure shear experiments. In figure 9 are plotted experimental
428 and identified Piola-Kirchhoff stresses versus principle stretch at equilibrium
429 for simple extension and pure shear. The relative error of the relation 60
430 was calculated for both experiments, its average value is 0,5 % for simple
431 extension and 2,3 % for pure shear which are very satisfactory considering
432 the non-linearity of the material.

433 6.2. Viscoelastic kernel

434 The identification of the viscoelastic kernel, as it's described in section 4,
435 is performed using two different experimental data: shear relaxation exper-
436 iment in the linear range of the behavior and dynamic tests for low level of
437 dynamic amplitude and without pre-strain. In what follows, results of this
438 identification procedure are discussed.

439 6.2.1. From shear relaxation experiment

440 The shear relaxation experiment is performed in simple shear deformation
441 at a strain level of 10 %. Despite that the value of the strain is quite big, it's
442 considered, however, in the linear range of the behavior since the material
443 is highly deformable. From figure 1 data for shear relaxation experiment
444 at 10 % are extracted and used in the identification of the Prony series
445 parameters. These parameters are reported in table 2. The average relative
446 error between the experimental and the identified viscoelastic kernel is in
447 the order of 0.1 %. The identified and the experimental normalized shear
448 relaxation modulus are plotted versus time in figure 10.

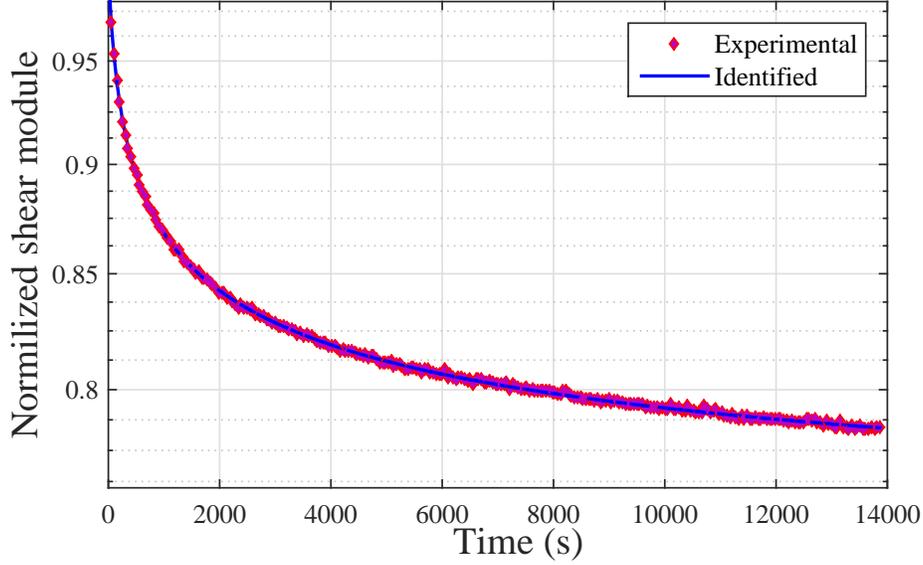


Figure 10: Normalized shear relaxation modulus of BIIR rubber versus time.

449 *6.2.2. From dynamic experiments*

450 The dynamic experiments are performed in simple shear deformation with
 451 a small dynamic amplitude and without a pre-strain of the form of equation
 452 45. It's recalled that the problem of the identification of the viscoelastic
 453 parameters from dynamic data of relation 48 is an ill-posed problem. Hence,
 454 a regularization procedure of Tikhonov is used. In what follows, this method
 455 is recalled and applied to theoretical dynamic data using parameters from
 456 [36] and then applied to dynamic data from [13].

- 457 • Tikhonov regularization method:

458 The linear system arising from the identification of the Prony series
 459 parameters from dynamic data is an ill-posed problem [31]. From the
 460 original system of equation 48 the following system arise:

$$Ax = b \tag{61}$$

461 in which A is the global matrix of the system to be calculated from
 462 49, b is the vector of experimental storage modulus and loss modulus
 463 vectors \hat{G}' and \hat{G}'' and x is the vector of the Prony series parameters
 464 $G_i, i = 1..N$. Tikhonov regularization method replaces system 61 by:

$$(A^t A + \mu I) x = A^t b \tag{62}$$

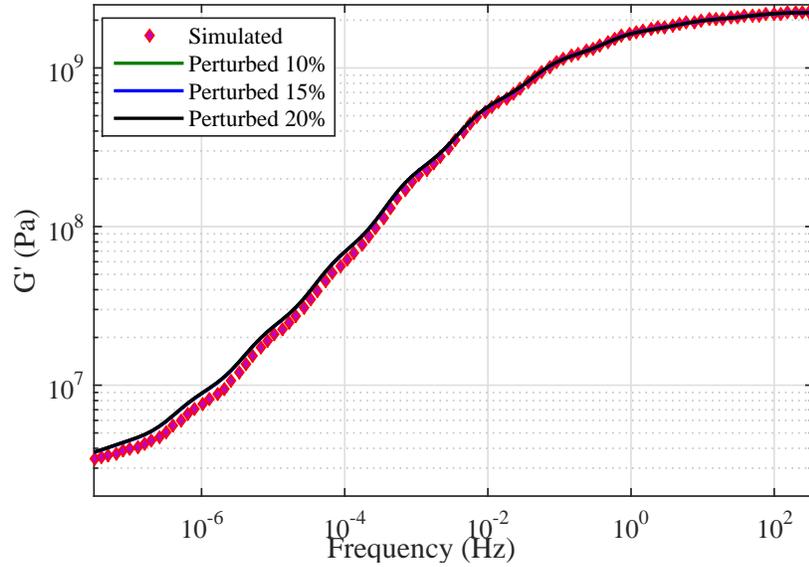
465 in which $\mu > 0$ is the regularization parameter and I is the identity
466 matrix. The regularization parameter is determined via a L-curve tech-
467 nique using Matlab software. It's well established that the solution of
468 system 62 noted x_μ gives the minimum residual for the minimization
469 problem arising from system 61 which means:

$$\begin{aligned} & \forall x \in \mathbb{R}^N \\ & \|Ax_\mu - b\|^2 \leq \|Ax - b\|^2, \text{ such that } \|x\|^2 \leq \|x_\mu\|^2 \end{aligned} \quad (63)$$

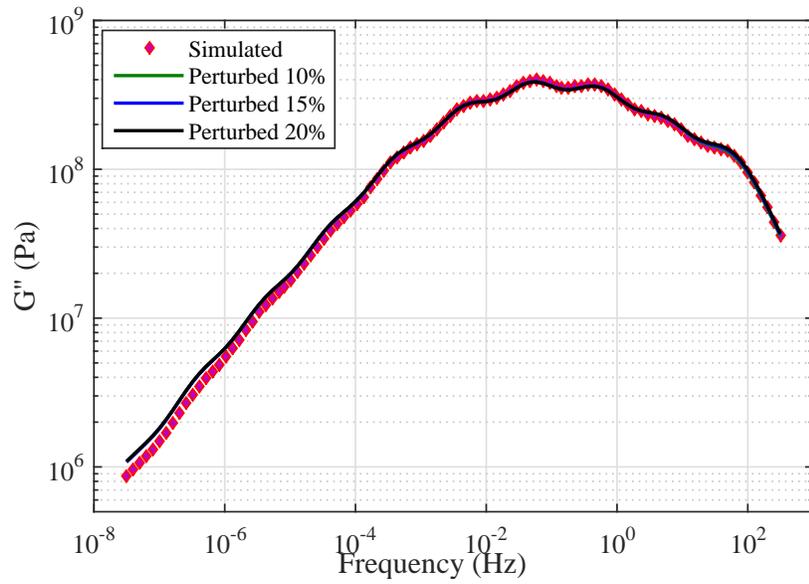
470 The proof of 63 and further development of the convergence of the
471 regularized Tikhonov problem are well studied in [37].

- 472 • Application of the Tikhonov method to simulated dynamic data:

473



(a) Storage modulus versus frequency for simulated and perturbed data



(b) Loss modulus versus frequency for simulated and perturbed data

Figure 11: Storage and loss moduli versus frequency for simulated and perturbed data

$G_i(Pa)$	$\tau_i(s)$
$1.94 \cdot 10^8$	$2 \cdot 10^{-2}$
$2.83 \cdot 10^8$	$2 \cdot 10^{-1}$
$5.54 \cdot 10^8$	$2 \cdot 10^0$
$6.02 \cdot 10^8$	$2 \cdot 10^1$
$3.88 \cdot 10^8$	$2 \cdot 10^2$
$1.56 \cdot 10^8$	$2 \cdot 10^3$
$4.1 \cdot 10^7$	$2 \cdot 10^4$
$1.38 \cdot 10^7$	$2 \cdot 10^5$
$3.68 \cdot 10^6$	$2 \cdot 10^6$
$7.9 \cdot 10^5$	$2 \cdot 10^7$
$9.6 \cdot 10^5$	$2 \cdot 10^8$

Table 3: Prony series parameters from [36]

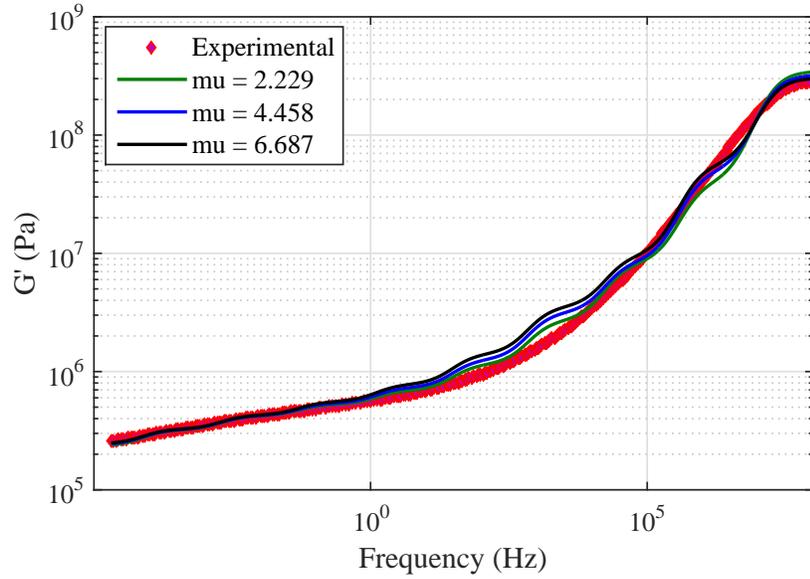
474 The Tikhonov regularization procedure described above was applied
475 to a simulated dynamic data generated using Prony series parameters
476 from [36] and relations in (47). Further, in order to test the capacity of
477 the method to deal with noisy experimental data, the second member
478 of system 62 was perturbed randomly as follow:

$$\tilde{b} = (1 \pm \varepsilon) b \quad (64)$$

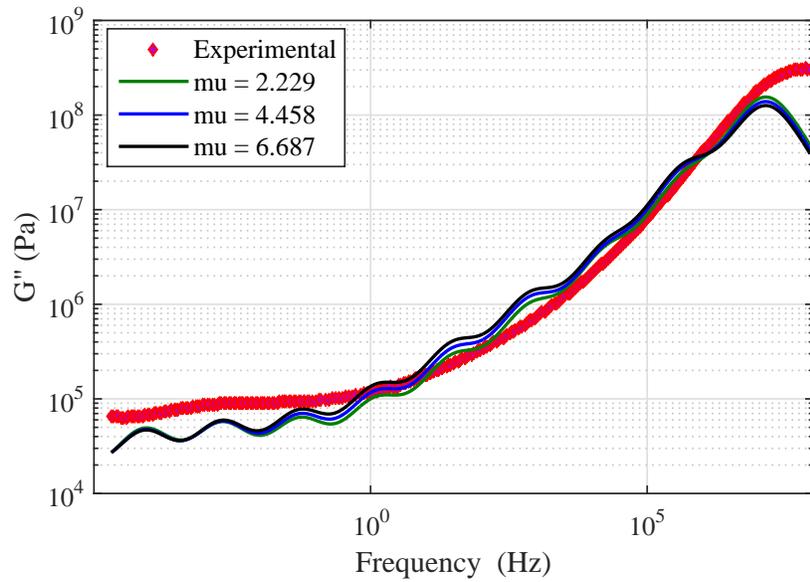
479 in which ε takes three different values: 10%, 15% and 20%. The Prony
480 series parameters from [36] are reported in table 3 with an equilibrium
481 modulus $G_\infty = 2.24 \cdot 10^6 Pa$. The results of this identification are re-
482 ported in figure 11 in terms of the dynamic moduli versus frequency
483 for the perturbed and original simulated data. The mean relative error
484 for the three perturbed data remains under 10% and hence this proce-
485 dure shows a huge capacity to predict the dynamic response functions
486 despite the perturbation of the second member of the system 63.

- 487 • Application of the Tikhonov method to dynamic data from [13]:
488 The dynamic experiment are represented by the storage and loss moduli
489 G' and G'' as functions of the frequency for the experimental frequency
490 window of $[10^{-5}, 10^7 Hz]$. The tikhonov regularization is applied and
491 a regularization parameter $\mu = 2.229$ is obtained for a 12 parameters
492 Prony series reported in table 4. The mean relative error is about

493 20%, this is due to the fact that the storage and loss moduli were
494 approximated via the WLF law [38] following the time-temperature
495 superposition principle.



(a) Storage modulus versus frequency for dynamic experimental data



(b) Loss modulus versus frequency for dynamic experimental data

Figure 12: Storage and loss moduli versus frequency for dynamic experimental data for different values of the regularization parameter μ

$G_i(Pa)$	$\tau_i(s)$
$3.13 \cdot 10^8$	$7.1 \cdot 10^{-8}$
$2.61 \cdot 10^7$	$1 \cdot 10^{-6}$
$9.86 \cdot 10^6$	$1.4 \cdot 10^{-5}$
$1.94 \cdot 10^6$	$1.98 \cdot 10^{-4}$
$9.42 \cdot 10^5$	$2.8 \cdot 10^{-3}$
$3.03 \cdot 10^5$	$3.92 \cdot 10^{-2}$
$1.4 \cdot 10^5$	$5.52 \cdot 10^{-1}$
$8.53 \cdot 10^4$	$7.77 \cdot 10^0$
$8.45 \cdot 10^4$	$1.09 \cdot 10^2$
$7.72 \cdot 10^4$	$1.53 \cdot 10^3$
$7.71 \cdot 10^4$	$2.16 \cdot 10^4$
$2.06 \cdot 10^4$	$3.05 \cdot 10^5$

Table 4: Prony series parameters from experimental dynamic data

496 *6.3. Reduced time function*

497 The reduced time is identified using the discretization formula 50 and
498 monotonic experiments of simple extension for two strain rates: $100\% s^{-1}$
499 and $200\% s^{-1}$. Cauchy stress versus principle stretch are plotted in figure
500 13. Results of the identification are reported in figure 14 in terms of the
501 reduced time coefficient which is a nonlinear function of time for the strain
502 rates considered. The pure shear experiment was predicted using the reduced
503 time, the predicted and experimental data for this experiment are plotted in
504 figure 15 against the principle stretch. From this result the relative error
505 of the Cauchy stress is calculated using relation 60, its mean value remains
506 under 2.5%. Hence, the proposed model is suitable to describe the material's
507 behavior at low and moderate strains.

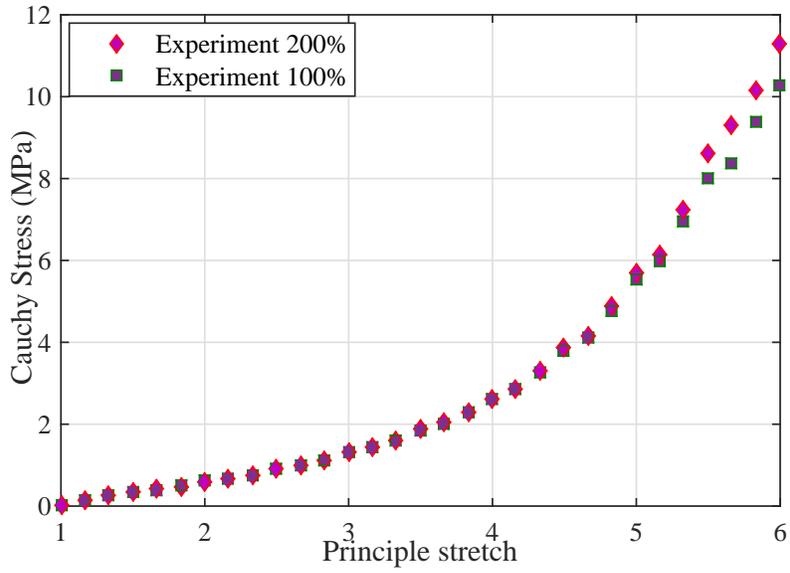


Figure 13: Cauchy stress versus principle stretch for simple extension experiment

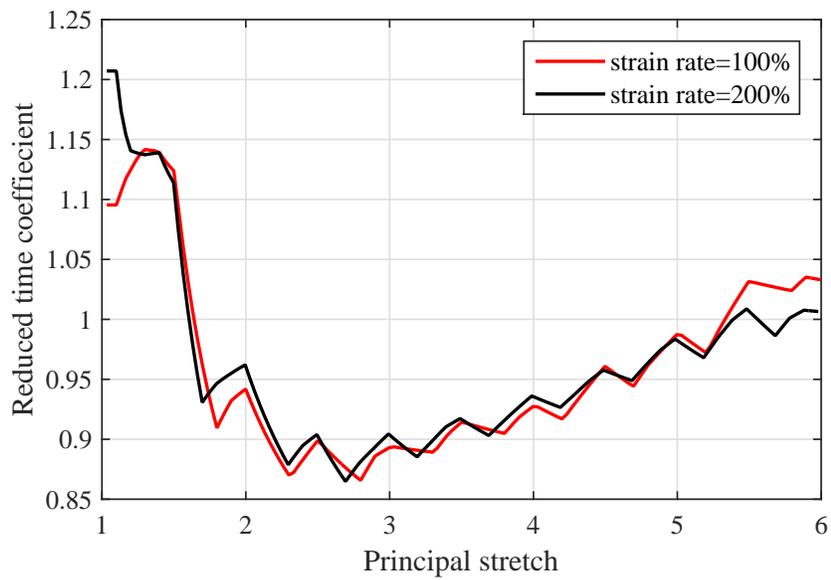


Figure 14: Reduced time coefficient for two strain rates $100\% s^{-1}$ and $200\% s^{-1}$

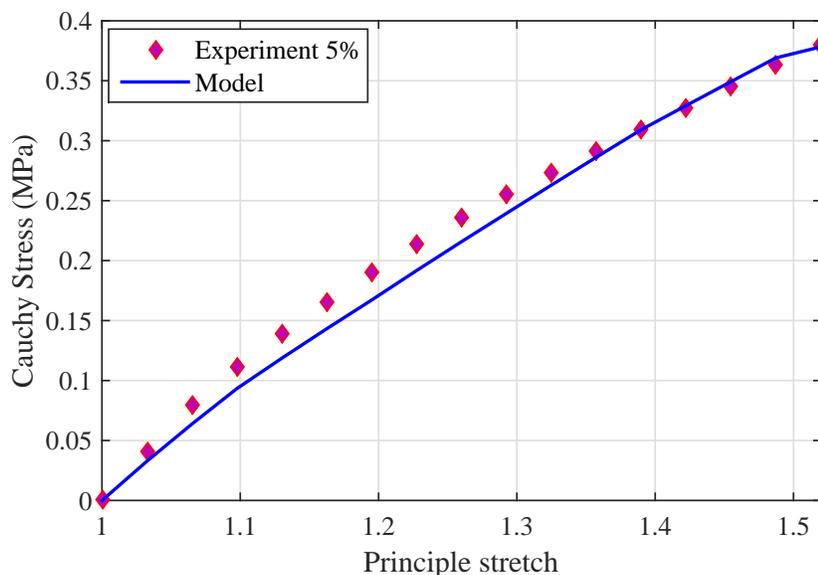


Figure 15: Cauchy stress versus principle stretch for pure shear experiment: experimental (diamond) model (solid curve)

508 7. Conclusion

509 A three-dimensional viscoelastic model at finite strain that incorporate a
510 strain dependent relaxation times has been proposed to describe nonfactoriz-
511 able behavior of rubber-like materials. The model is based upon the internal
512 state variables approach and the framework of rational thermodynamics and
513 experimental arguments. The free energy density is decomposed into a vol-
514 umetric and deviatoric parts. Furthermore, thermodynamic restrictions are
515 fulfilled via a sufficient condition on the model's parameters resulting from
516 the application of the Clausius-Duhem inequality for an arbitrary process.
517 The identification of several functions involved in the model was, then, ad-
518 dressed. Each function's identification procedure turns out to the resolution
519 of a linear or nonlinear system. Moreover, a regularization procedure of
520 Tikhonov was applied in the resolution of the ill-posed problem arising from
521 the identification of the viscoelastic kernel from dynamic data.
522 This identification procedure was applied to a generated data from a multi-
523 integral viscoelastic model and a static and dynamic experimental charac-
524 terization of a BIIR rubber. The results of the identification has shown a
525 huge capacity of the model to describe both experimental and multi-integral

526 viscoelastic model in the time domain of the behavior within and outside the
527 range of strain rates considered in deriving the model.
528 Our main object was to develop a constitutive equation compatible with the
529 second law of thermodynamics and suitable with the Finite-elements theory.
530 The implementation of the model within Abaqus software and the simulation
531 of real component with complex industrial application is the goal of future
532 work.

533 **References**

- 534 [1] R. A. Schapery, An engineering theory of nonlinear viscoelasticity with
535 applications, *International Journal of Solids and Structures* 2 (1966)
536 407–425.
- 537 [2] W. Chang, R. Bloch, N. Tschoegl, Time-dependent response of soft
538 polymers in moderately large deformations, *Proceedings of the National
539 Academy of Sciences* 73 (1976) 981–983.
- 540 [3] W. Chang, R. Bloch, N. Tschoegl, Study of the viscoelastic behavior of
541 uncrosslinked (gum) rubbers in moderately large deformations, *Journal
542 of Polymer Science: Polymer Physics Edition* 15 (1977) 923–944.
- 543 [4] J. Sullivan, A nonlinear viscoelastic model for representing nonfactor-
544 izable time-dependent behavior in cured rubber, *Journal of Rheology
545 (1978-present)* 31 (1987) 271–295.
- 546 [5] R. A. Schapery, On the characterization of nonlinear viscoelastic mate-
547 rials, *Polymer Engineering & Science* 9 (1969) 295–310.
- 548 [6] K. C. Valanis, Thermodynamics of large viscoelastic deformations, *Stud-
549 ies in Applied Mathematics* 45 (1966) 197–212.
- 550 [7] W. G. Knauss, I. Emri, Volume change and the nonlinearly thermo-
551 viscoelastic constitution of polymers, *Polymer Engineering & Science
552* 27 (1987) 86–100.
- 553 [8] J. M. Caruthers, D. B. Adolf, R. S. Chambers, P. Shrikhande, A ther-
554 modynamically consistent, nonlinear viscoelastic approach for modeling
555 glassy polymers, *Polymer* 45 (2004) 4577–4597.

- 556 [9] R. A. Simo, On a fully three-dimensional finite-strain viscoelastic damage
557 model: Formulation and computational aspects, *Computer Methods*
558 *in Applied Mechanics and Engineering* 60 (1987) 153–173.
- 559 [10] G. Holzapfel, R. A. Simo, A new viscoelastic constitutive model for con-
560 tinuous media at finite thermomechanical changes, *International Journal*
561 *of Solids and Structures* 33 (1996) 3019–3034.
- 562 [11] S. Reese, S. Govindjee, A theory of viscoelasticity and numerical ap-
563 plications, *International Journal of Solids and Structures* 35 (1998)
564 3455–3482.
- 565 [12] A. C. Pipkin, Small finite deformations of viscoelastic solids, *Reviews*
566 *of Modern Physics* 36 (1964) 1034–1041.
- 567 [13] J. Nidhal, Characterization and comparison of viscoelastic properties of
568 three vulcanized rubber materials for a damping application: Natural
569 rubber, bromobutyl and a natural rubber/bromobutyl mixture (????).
- 570 [14] E. Peña, J. A. Peña, M. Doblar, On modelling nonlinear viscoelastic
571 effects in ligaments, *Journal of Biomechanics* 41 (2008) 2659–2666.
- 572 [15] P. O’connell, G. McKenna, Large deformation response of polycarbon-
573 ate: Time-temperature, time-aging time, and time-strain superposition,
574 *Polymer Engineering & Science* 37 (1997) 1485–1495.
- 575 [16] N. W. Tschoegl, W. G. Knauss, I. Emri, The effect of temperature
576 and pressure on the mechanical properties of thermo-and/or piezorhe-
577 ologically simple polymeric materials in thermodynamic equilibrium—a
578 critical review, *Mechanics of Time-Dependent Materials* 6 (2002) 53–99.
- 579 [17] J. C. Simo, T. J. R. Hughes, *Computational inelasticity*, volume 7,
580 Springer Science & Business Media, 2006.
- 581 [18] N. W. Tschoegl, Time dependence in material properties: An overview,
582 *Mechanics of Time-Dependent Materials* 1 (1997) 3–31.
- 583 [19] S. Matsuoka, C. J. Aloisio, H. E. Bair, Interpretation of shift of re-
584 laxation time with deformation in glassy polymers in terms of excess
585 enthalpy, *Journal of Applied Physics* 44 (1973) 4265–4268.

- 586 [20] P. J. Flory, Thermodynamic relations for high elastic materials, Trans-
587 actions of the Faraday Society 57 (1961) 829–838.
- 588 [21] G. A. Holzapfel, J. C. Simo, A new viscoelastic constitutive model
589 for continuous media at finite thermomechanical changes, International
590 Journal of Solids and Structures 33 (1996) 3019–3034.
- 591 [22] B. D. Coleman, M. E. Gurtin, Thermodynamics with internal state
592 variables, The Journal of Chemical Physics 47 (1967) 597–613.
- 593 [23] C. Truesdell, W. Noll, The non-linear field theories of mechanics, vol-
594 ume 3, Springer, 2004.
- 595 [24] V. Shim, L. Yang, C. Lim, P. Law, A visco-hyperelastic constitutive
596 model to characterize both tensile and compressive behavior of rubber,
597 Journal of Applied Polymer Science 92 (2004) 523–531.
- 598 [25] L. Yang, V. Shim, A visco-hyperelastic constitutive description of elas-
599 tomeric foam, International journal of impact engineering 30 (2004)
600 1099–1110.
- 601 [26] R. Rivlin, Large elastic deformations of isotropic materials. iv. further
602 developments of the general theory, Philosophical Transactions of the
603 Royal Society of London A: Mathematical, Physical and Engineering
604 Sciences 241 (1948) 379–397.
- 605 [27] E. Pucci, G. Saccomandi, A note on the gent model for rubber-like
606 materials, Rubber chemistry and technology 75 (2002) 839–852.
- 607 [28] R. Ogden, G. Saccomandi, I. Sgura, Fitting hyperelastic models to
608 experimental data, Computational Mechanics 34 (2004) 484–502.
- 609 [29] N. Tschoegl, I. Emri, Generating line spectra from experimental re-
610 sponses. part ii: Storage and loss functions, Rheologica Acta 32 (1993)
611 322–327.
- 612 [30] W. G. Knauss, J. Zhao, Improved relaxation time coverage in ramp-
613 strain histories, Mechanics of Time-Dependent Materials 11 (2007) 199–
614 216.

- 615 [31] C. Elster, J. Honerkamp, J. Weese, Using regularization methods for
616 the determination of relaxation and retardation spectra of polymeric
617 liquids, *Rheologica acta* 31 (1992) 161–174.
- 618 [32] M. Nashed, The theory of tikhonov regularization for fredholm equations
619 of the first kind (cw groetsch), *SIAM Review* 28 (1986) 116–118.
- 620 [33] D. Hibbit, B. Karlsson, P. Sorensen, *Abaqus/theory manual*, Rhode
621 Island 6 (2007).
- 622 [34] S. Hassani, A. A. Soulimani, A. Ehrlacher, A nonlinear viscoelas-
623 tic model: the pseudo-linear model, *European Journal of Mechanics-*
624 *A/Solids* 17 (1998) 567–598.
- 625 [35] R. W. Ogden, G. Saccomandi, I. Sgura, Fitting hyperelastic models to
626 experimental data, *Computational Mechanics* 34 (2004) 484–502.
- 627 [36] S. W. Park, R. A. Schapery, Methods of interconversion between linear
628 viscoelastic material functions. part ia numerical method based on prony
629 series, *International Journal of Solids and Structures* 36 (1999) 1653–
630 1675.
- 631 [37] D. Calvetti, S. Morigi, L. Reichel, F. Sgallari, Tikhonov regularization
632 and the l-curve for large discrete ill-posed problems, *Journal of Compu-*
633 *tational and Applied Mathematics* 123 (2000) 423 – 446.
- 634 [38] M. L. Williams, R. F. Landel, J. D. Ferry, The temperature depen-
635 dence of relaxation mechanisms in amorphous polymers and other glass-
636 forming liquids, *Journal of the American Chemical society* 77 (1955)
637 3701–3707.