Visco-hyperelastic model with internal state variable coupled with discontinuous damage concept under total Lagrangian formulation

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Abstract

The silica-filled rubber material presented in this paper exhibits nonlinear elasticity, nonlinear rate dependence and stress-softening effect under cyclic loading. In order to model the material behavior in a finite element code, the internal state variable concept is considered for finite deformation viscoelasticity. Moreover, the so-called Mullins’ effect is taken into account by using a discontinuous damage concept. A total Lagrangian formulation with incompressibility constraint is adopted in the finite element code. The constitutive equations with their optimized set of parameters are validated by comparing the simulated results with experimental data. This result is very useful for the fatigue lifetime analysis of the investigated silica-filled rubber material.

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1. Introduction

The present work is part of a general study on the fatigue lifetime of a filled rubber material used in shoe-sole manufacturing. The scientific approach followed for this analysis is to derive a diagram enabling to predict the number of cycles to crack initiation. This diagram should display on its axes relevant parameters which have to be calculated at every location of the prescribed structure. This task requires a tool that is capable of computing, for every loaded sample, not only the global

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parameters (i.e. measurable parameters) but also local quantities such as stress and strain tensors. This is obtained with the help of a finite element (FE) code.

The reliability on a FE code essentially depends on the provided material constitutive equations, which are determined by using experimental database on simple specimens. The material of interest exhibits nonlinear elasticity, stress-softening effect under cyclic loading and a nonlinear rate dependence. These three main components of the mechanical behavior are addressed in this paper.

The first section presents the silica filled rubber material as well as the experimental procedures used to analyse the constitutive equations.

The constitutive equations based upon internal state variable (ISV) are then described in detail. The basis of the theory (Sidoroff, 1975a) deals with a tensorial ISV concept. In addition, the evolution equation is assumed to be governed by a dissipation pseudo-potential.

The stress-softening effect (Mullins’ effect) model utilizes the discontinuous damage concept (Miehe, 1995).

Finally, FE results are shown by outlining the optimization of the set of material parameters, the simulation of the simple tests and the validation of the optimized set of parameters on more complex geometries in 2D and 3D conditions.

2. Material and experimental procedures

2.1. The silica filled styrene butadiene rubber material

The material of interest is a silica filled styrene butadiene rubber (SBR). Scanning electron microscopy (SEM) examinations carried out on a microtomed slice, show that the microstructure is strongly filled with silica aggregates (1–5 μm) and silica agglomerates (10–100 μm). Note that the silica is added to the compound formulation in order to improve the wear resistance of the material. According to chemical analyses, the silica filler concentration is about 30% (wt.%). Furthermore, local wavelength dispersive spectrometry (WDS) analysis indicates that these fillers are in fact silica-particles-occluded-gum clusters. We should also mention the presence of carbon black fillers.

2.2. The experimental procedures

Detailed experimental results have been reported (Robisson, 2000). In this paper, we address two categories of mechanical tests:

- Experimental database for material coefficients optimization. The material of interest exhibits three main components of mechanical behavior. Let us consider first uniaxial cyclic loading performed on rectangular strips (20×150 mm) cut from calandered sheets of 3 mm thickness. The loading sequence is imposed with displacement control, whereas the unloading sequence is load-controlled to zero in order to prevent buckling of the specimen. Fig. 1 displays
the engineering stress versus elongation curve of a test performed at $\lambda_{\text{max}} = 1.5$. In this section, we are only interested in experimental data (solid line). The FE simulation (dashed line) will be addressed further. All of the aforementioned three aspects of the mechanical behavior appear in this figure. The hyperelasticity is characterized by the S-shaped curve from the second loading on. The Muffins’ effect makes the first loading path to differ from the second and the unloading paths. Finally, the viscoelasticity imposes, on the one hand, the stress relaxation at $\lambda_{\text{max}}$, on the other hand, the progressive elongation set at zero load. Note that non—linear time-dependent behavior of polymers in general has been experimentally demonstrated by authors like Khan and Zhang (2001) or Khan and Lopez-Pamies (in press). For the rubber material of interest and in order to uncouple all of the aforementioned intermixed three components, further tests have been carried out:

- Monotonic uniaxial tension and compression tests for hyperelasticity.
- Stress relaxation tests under uniaxial tension and compression for viscoelasticity. The loading is applied with the maximum strain rate, the stress–strain curve obtained from these tests is assumed to be “viscosity-free” and hence can be supposed to be the curve of first stretch to analyze the Mullins’ effect. Additionally, the stress decay during the relaxation test allows to determine the relaxation times of the material.
- Cyclic uniaxial tension/tension tests with various $\lambda_{\text{max}}$ for all of the three aspects. Note that Mullins’ effect stabilized curve has been studied on this type of test since the second and further loading sequences reduce to a unique stress–strain curve different from the one of the first stretch.

- The experimental database for validation purpose consists of tests performed on axisymmetrically notched specimens. These tests aim at determining the
number of cycles to crack initiation (1 mm size optically detectable). Even if the fatigue lifetime is out of the scope of this paper, the experimental database is interesting from a numerical simulation standpoint because these tests may validate the constitutive equations and the set of material parameters may be optimized thanks to the above test results. For cyclic tension/compression tests, the notch root radius is about 40 mm (Fig. 2 left). Henceforth, this specimen will be referred to as NS40 specimen. Whereas for cyclic torsional tests, specimens with a notch radius of about 2 mm (Fig. 2, right) (henceforth NS2) are used.

3. Constitutive equations

3.1. Thermodynamic state variables

All of the tests were carried out at room temperature. The cyclic tests were performed at low frequencies in order to avoid self-heating of the sample. For these reasons, the transformation is supposed to be isothermal. In other words, the thermal aspects will not be considered in the following.

Let \( \mathbf{F} \) be the deformation gradient relating the transformation from the reference configuration \( C_0 \) to the current configuration \( C_t \). The right Cauchy–Green tensor corresponding to \( \mathbf{F} \) is \( \mathbf{C} = \mathbf{F}^T \mathbf{F} \). The second Piola–Kirschhoff stress tensor is denoted by \( \mathbf{S} \).

Let us start with the Clausius–Duhem inequality, by assuming no heat exchange hypothesis, and by considering only the intrinsic dissipation.

\[
\left( \mathbf{S} - 2 \frac{\partial W}{\partial \mathbf{C}} \right) : \dot{\mathbf{C}} - 2 \frac{\partial W}{\partial \mathbf{A}_i} \dot{\mathbf{A}}_i \geq 0
\]

(1)

Fig. 2. Axisymmetrically notched specimens (NS40 left and NS2 right) for cyclic tension/compression and torsional tests
where: $W$ is the Helmholtz free energy; $A_i$ is an internal variable in the reference configuration.

Eq. (1) can be written as follows:

$$
S_v : \dot{C} + W_i : \dot{A}_i \geq 0
$$

where:

$$
S_v = S - 2 \frac{\partial W}{\partial C}
$$

$$
W_i = -2 \frac{\partial W}{\partial A_i}
$$

In order to write the evolution equation, it is postulated (Sidoroff, 1975a) that a dissipation pseudo-potential $\phi$ exists. Germain (1986) characterized this dissipation pseudo-potential $\phi$ by considering the thermodynamics of the non-reversible dissipative processes with internal variables. $\phi$ relates the thermodynamical forces ($S_v$, $W_i$) to their corresponding fluxes ($\dot{C}$, $\dot{A}_i$).

$$
S_v = 2 \frac{\partial \phi}{\partial C}
$$

$$
W_i = 2 \frac{\partial \phi}{\partial A_i}
$$

3.2. Intermediate configuration

The extension of an ISV theory to the 3D case imposes to the tensorial ISV to be related to an “intermediate configuration” (Sidoroff, 1975a; Carpentier-Gabrieli, 1995; Lion, 1997; Reese and Govindjee, 1998; Huber and Tsakmakis, 2000). This is obtained by adopting the multiplicative split of the deformation gradient $F = F^e F^v$, where $F^e$ (superscript $e$ stands for “viscoelastic”) deals with the $C_0$ to the intermediate configuration $C_i$ transformation and $F^v$ (superscript $v$ stands for “elastic”) relates the $C_i$ to $C_t$ transformation.

Let $C^e = F^{eT} F^e$ and $C^v = F^{vT} F^v$ be the right Cauchy–Green strain tensors respectively associated to the elastic and viscoelastic, deformation gradients $F^e$ and $F^v$.

The relation $F = F^e F^v$ imposes the choice of two (free) tensorial variables among the three corresponding right Cauchy–Green strain tensors. The first one is obviously $C$ since it represents the input variable. By analogy with the small strain theory where the dissipation pseudo-potential is a function of the viscoelastic strain rate, the choice of the tensorial ISV will refer to the rate of the “viscoelastic” right Cauchy–Green tensor $\dot{C}^v$. Accordingly, the adopted couple of thermodynamic tensorial variables is $(C, \dot{C}^v)$. 
Eqs. (3) and (5) yield:

\[ S = 2 \frac{\partial W}{\partial C} + 2 \frac{\partial \phi}{\partial C} \]  

(7)

whereas Eqs. (4) and (6) lead to:

\[ - \frac{\partial W}{\partial C_v} = \frac{\partial \phi}{\partial C_v} \]  

(8)

Note that Eqs. (7) and (8) require isotropy assumption (Carpentier-Gabrieli, 1995).

The incompressibility constraint of the material implies the introduction of a Lagrange multiplier \( p \) in such a way that the Cauchy stress tensor is defined by adding a pressure term \( p I \). This latter is transformed to its total Lagrangian counterpart by the expression \( p C^{-1} \). For the global incompressibility constraint acting on \( C \) the term \( p C^{-1} \) has to be added to the right part of Eq. (7). By considering the incompressibility of the \( C_v \) ISV, a second Lagrange multiplier \( q \) is added allowing to introduce \( q C_v^{-1} \) to the evolution equation [Eq. (8)].

Therefore, Eqs. (7) and (8) become:

\[ S = 2 \frac{\partial W}{\partial C} + 2 \frac{\partial \phi}{\partial C} - p C^{-1} \]  

(9)

\[ \frac{\partial \phi}{\partial C_v} + \frac{\partial W}{\partial C_v} - q C_v^{-1} = 0 \]  

(10)

3.3. Rheological models

According to Sidoroff (1975b), various rheological models are available. Let us focus upon the model for a material capable of straining instantaneously (non viscous material), creeping and stress relaxing.

The simplest models of this kind are what Huber and Tsakmakis (2000) call the “Three parameter solids” models. The first one is a spring in series with a Kelvin element (Fig. 3): the Poynting–Thomson rheological model. The second one consists of a spring in parallel with a Maxwell element: the Zener model. Both models are equivalent in the small strain case (linear viscoelasticity). This is no longer true for their extension to finite deformation: in fact, the two models do not predict the same mechanical response (Carpentier-Gabrieli, 1995; Huber and Tsakmakis, 2000; Andrieux, 1996).

Lion (1996) noted that the Zener model refers to an equilibrium elastic state corresponding to the creep stabilized material (infinitely low strain rate, \( t \to \infty \)). Comparatively, the Poynting–Thomson model is based on the instantaneous elasticity (infinitely high strain rate, \( t \to 0 \)).
It should be mentioned that Zener’s model is often developed in the literature.

- Programming Zener’s model in order to implement it in a finite element code is more convenient due to the relative simplicity of its equations.
- It is easy to extend Zener’s model to several spring/dashpot branches in parallel corresponding to several relaxation times, due to its basic architecture (already in parallel).

Both Poynting–Thomson and Zener models can be developed within the formalism adopted in this paper. Nevertheless, only the Poynting–Thomson will be presented in the remainder of the paper. In our opinion, this latter is not so frequently addressed as the Zener’s model in the literature.

3.3.1. The polynomial deformation energies

In the Poynting–Thomson rheological model (Fig. 3), the total deformation energy can be split into two parts \( W = W_e(\mathbf{C}^e) + W_v(\mathbf{C}^v) \), whereas the dissipation pseudo-potential can be expressed as \( \phi(\mathbf{C}^v) \). \( W_e \) and \( W_v \) are respectively related to the springs in the “elastic” and “viscous” branches. Note that \( \phi \) only depends on \( \mathbf{C}^v \), therefore, in Eq. (9), the derivative \( \partial \phi / \partial \mathbf{C} \) disappears.

The classical Rivlin deformation energy is proposed in this paper:

\[
W_e = \sum_{i+j=1}^{n} c_{ij}^{e} (I_1^e - 3) ^i (I_2^e - 3) ^j \\
W_v = \sum_{i+j=1}^{m} c_{ij}^{v} (I_1^v - 3) ^i (I_2^v - 3) ^j
\]

\( c_{ij}^{e} \) and \( c_{ij}^{v} \) are material parameters, \((I_1^e, I_2^e)\) (resp. \((I_1^v, I_2^v)\)) are the first and second invariants of the “elastic” (resp. “viscoelastic”) right Cauchy–Green tensors. In fact, for a \( \mathbf{C} \) tensor \( I_1 = \text{trace}(\mathbf{C}) \) and \( I_2 = \frac{1}{2} (I_1^2 - \text{trace}(\mathbf{C} \cdot \mathbf{C})) \).

Fig. 3. Poynting–Thomson rheological model.
The deformation energy $W_e$ is function of the elastic right Cauchy–Green tensor $C^e$. The couple of state variables being $(C, C^v)$. The multiplicative split of the deformation gradient allows to express $W_e$ as a function of $C$ and $C^v$.

It should be noted that an hyperelastic model of the network polymer chains can be used instead of this phenomenological Rivlin polynomial deformation energy (see for example Sweeney et al., 2002).

3.3.2. The viscoelastic evolution equation

In order to have a full description of the visco-hyperelasticity set of equations, one has to give an expression for the dissipation pseudo-potential $\phi$. Following Boukamel et al. (1997):

$$\phi = \frac{1}{2} f_v \dot{C}^v \cdot \dot{C}^v$$

where $f_v$ is a “viscosity” coefficient, hence:

$$\frac{\partial \phi}{\partial C^v} = f_v \dot{C}^v$$

3.4. The basic equations

With respect to the Poynting–Thomson rheological model, the set of two state equations [Eqs. (9) and (10)] can be rewritten as:

$$S = 2 \frac{\partial W_e}{\partial C} - \rho C^{-1}$$

$$\frac{\partial \phi}{\partial C^v} + \frac{\partial W_e}{\partial C^v} + \frac{\partial W_v}{\partial C^v} - q C^v \cdot C^{-1} = 0$$

In the following, we seek to calculate each expression for the partial derivative of the deformation energy with respect to the right Cauchy–Green tensor. By transforming Eqs. (15) and (16), we write the initial set of equations from which we will start the development:

$$S = 2 \frac{\partial W_e}{\partial I_1^e} \frac{\partial I_1^e}{\partial C} + 2 \frac{\partial W_e}{\partial I_2^e} \frac{\partial I_2^e}{\partial C} - \rho C^{-1}$$

$$\frac{\partial \phi}{\partial C^v} + \frac{\partial W_e}{\partial I_1^e} \frac{\partial I_1^e}{\partial C^v} + \frac{\partial W_e}{\partial I_2^e} \frac{\partial I_2^e}{\partial C^v} + \frac{\partial W_v}{\partial I_1^v} \frac{\partial I_1^v}{\partial C^v} + \frac{\partial W_v}{\partial I_2^v} \frac{\partial I_2^v}{\partial C^v} - q C^v \cdot C^{-1} = 0$$

The proofs for all equations displayed in the next two subsections are detailed in the Appendix.
3.4.1. Rearrangement of the first equation: constitutive law

The expressions of the partial derivatives of the “elastic” first and second invariants with respect to the total right Cauchy–Green tensor are (see the Appendix for the details):

\[
\frac{\partial I^e_1}{\partial \mathbf{C}} = \mathbf{C}^{-1} \quad (19)
\]

\[
\frac{\partial I^e_2}{\partial \mathbf{C}} = I^e_1 \mathbf{C}^{-1} - \mathbf{C}^{-1} - \mathbf{C} \mathbf{C}^{-1} \quad (20)
\]

Accordingly, the final expression of the constitutive law [Eq. (17)] is:

\[
\mathbf{S} = 2 \left( \left( \frac{\partial W_e}{\partial I^e_1} + \frac{\partial W_e}{\partial I^e_2} I^e_1 \right) \mathbf{I} - \frac{\partial W_e}{\partial I^e_2} \mathbf{C} \mathbf{C}^{-1} \mathbf{C} \right) \mathbf{C}^{-1} - \rho \mathbf{C}^{-1} \quad (21)
\]

3.4.2. Rearrangement of the second equation: the evolution law

The partial derivatives of the “elastic” first and second invariants with respect to the “viscoelastic” right Cauchy–Green tensor are expressed by (see the Appendix for the details):

\[
\frac{\partial I^e_1}{\partial \mathbf{C}^v} = -\mathbf{C}^{-1} \mathbf{C} \mathbf{C}^{-1} \quad (22)
\]

\[
\frac{\partial I^e_2}{\partial \mathbf{C}^v} = -I^e_1 \mathbf{C}^{-1} \mathbf{C} \mathbf{C}^{-1} + \mathbf{C}^{-1} \left( \mathbf{C} \mathbf{C}^{-1} \mathbf{C} \right) \mathbf{C}^{-1} \quad (23)
\]

Moreover,

\[
\frac{\partial I^v_1}{\partial \mathbf{C}^v} = \mathbf{I} \quad (24)
\]

\[
\frac{\partial I^v_2}{\partial \mathbf{C}^v} = I^v_1 \mathbf{I} - \mathbf{C} \quad (25)
\]

The final expression of the evolution equation is then:

\[
\frac{\partial \mathbf{\phi}}{\partial \mathbf{C}^v} - \mathbf{C}^{-1} \mathbf{C} \left( \frac{\partial W_e}{\partial I^e_1} \mathbf{I} + \frac{\partial W_e}{\partial I^e_2} \left( I^e_1 \mathbf{I} - \mathbf{C} \mathbf{C}^{-1} \mathbf{C} \right) \right) \mathbf{C}^{-1} + \frac{\partial W_v}{\partial I^v_1} \mathbf{I} + \frac{\partial W_v}{\partial I^v_2} \left( I^v_1 \mathbf{I} - \mathbf{C} \right) - q \mathbf{C}^{-1} = 0 \quad (26)
\]

or, equivalently:
\[ f_v \ddot{C} - C^{v-1} C \left( \left( \frac{\partial W_v}{\partial I_1^e} + \frac{\partial W_v}{\partial I_2^e} I_1 \right) - \frac{\partial W_v}{\partial I_2^e} C^{v-1} C \right) C^{v-1} \]
\[ + \left( \frac{\partial W_v}{\partial I_1^e} + \frac{\partial W_v}{\partial I_2^e} I_1 \right) I - \frac{\partial W_v}{\partial I_2^e} C^{v} - q C^{v-1} = 0 \]  
\( (27) \)

3.5. Computational strategy

The evolution law [Eq. (27)] is a differential equation on \( C^v \).
The input variable is the deformation gradient tensor \( F^v \). The right Cauchy–Green tensor computation is straightforward \( C = F^T F \).

Let \( C^v \) be the ISV:
\[
C^v = \begin{bmatrix}
C^v_{11} & C^v_{12} & C^v_{13} \\
C^v_{12} & C^v_{22} & C^v_{23} \\
C^v_{13} & C^v_{23} & C^v_{33}
\end{bmatrix}
\]  
\( (28) \)

\( C^v \) must satisfy the following conditions:
- Initial state
\[
C^v(t = 0) = I
\]  
\( (29) \)

- Incompressibility applied to \( C^v \)
\[
\det(C^v) = 1
\]  
\( (30) \)

From Eq. (28), the \( \dot{C}^v \) tensor is:
\[
\dot{C}^v = \begin{bmatrix}
\dot{C}^v_{11} & \dot{C}^v_{12} & \dot{C}^v_{13} \\
\dot{C}^v_{12} & \dot{C}^v_{22} & \dot{C}^v_{23} \\
\dot{C}^v_{13} & \dot{C}^v_{23} & \dot{C}^v_{33}
\end{bmatrix}
\]  
\( (31) \)

\( \dot{C}^v \) terms have to satisfy Eq. (30):
\[
\frac{\partial \left( \det(C^v) \right)}{\partial t} = 0
\]  
\( (32) \)

Note that the initial state [Eq. (29)] of \( C^v \) induces the incompressibility constraint to be held during the whole transformation.

Expanding the evolution equation leads to a set of 6 differential equations with 6 unknowns: \( (C^v_{11}, C^v_{22}, C^v_{33}, C^v_{12}, C^v_{23}, C^v_{13}) \). The Lagrange multiplier \( q \) can be eliminated by utilizing either Eq. (30) or both Eqs. (32) and (29).
Finally, the set of differential equations can be summarized as follows:

\[ \dot{\mathbf{C}}_{ij}^{v} = \mathbf{F}_k \left( \mathbf{C}_{pr}^{v}, \mathbf{F}_{mn} \right) \text{ where } (i,j,p,r,m,n) \in \{1,2,3\} \text{ and } 1 \leq k \leq 6 \]  

(33)

This set of equations is integrated numerically (using a fourth order Runge–Kutta scheme), which gives access to the \( \mathbf{C}^{v} \) history. The second Piola–Kirchoff (PK2) stress tensor is then derived in a straightforward manner by utilizing the constitutive equation [Eq. (21)].

3.6. Worked examples

3.6.1. Worked example 1: uniaxial relaxation test

Let \( \mathbf{F} \) be the uniaxial deformation gradient taking into account the incompressibility

\[
\mathbf{F} = \begin{bmatrix}
\lambda & 0 & 0 \\
0 & 1/\sqrt{\lambda} & 0 \\
0 & 0 & 1/\sqrt{\lambda}
\end{bmatrix}
\]  

(34)

where \( \lambda \) is the axial stretch ratio. For the relaxation test, the material is supposed to be stretched up to \( \lambda = 2 \) with a stretch rate of 1/s. Then, this stretch is held at 2 for the remainder of the test (10 s in this example).

\[
\lambda = \begin{cases} 
1 + t & \text{if } t < 1 \\
2 & \text{otherwise}
\end{cases}
\]  

(35)

Let us define the \( \mathbf{C}^{v} \) tensor:

\[
\mathbf{C}^{v} = \begin{bmatrix}
\mathbf{C}_{11}^{v} & 0 & 0 \\
0 & 1/\sqrt{\mathbf{C}_{11}^{v}} & 0 \\
0 & 0 & 1/\sqrt{\mathbf{C}_{11}^{v}}
\end{bmatrix}
\]  

(36)

Note that this tensor obeys the incompressibility constraint \( \det(\mathbf{C}^{v}) = 1 \). The \( q \) parameter is eliminated by imposing \( C_{11}^{v}(t = 0) = 1 \).

\[
\dot{\mathbf{C}}^{v}_{11} = \begin{bmatrix}
\dot{C}_{11}^{v} & 0 & 0 \\
0 & -\dot{C}_{11}^{v}/(2\mathbf{C}_{11}^{v}\sqrt{\mathbf{C}_{11}^{v}}) & 0 \\
0 & 0 & -\dot{C}_{11}^{v}/(2\mathbf{C}_{11}^{v}\sqrt{\mathbf{C}_{11}^{v}})
\end{bmatrix}
\]  

(37)

The only differential equation on \( \mathbf{C}_{11}^{v} \) is integrated numerically (Runge–Kutta method). In order to go further, we have to provide with the material parameters.
The Rivlin strain energy for the first spring.

\[ W_e = c_{10}^e (I_1^e - 3) + c_{01}^e (I_2^e - 3) + c_{20}^e (I_2^e - 3)^2 \]

\[
\begin{align*}
&c_{10}^e (\text{MPa}) = 3.565 \\
&c_{01}^e (\text{MPa}) = 1.305 \\
&c_{20}^e (\text{MPa}) = 0.839
\end{align*}
\]

The Mooney–Rivlin strain energy for the second spring

\[ W_v = c_{10}^v (I_1^v - 3) + c_{01}^v (I_2^v - 3) \]

\[
\begin{align*}
&c_{10}^v (\text{MPa}) = 3.470 \\
&c_{01}^v (\text{MPa}) = 1.866
\end{align*}
\]

The “viscosity” coefficient \( f_v = 47 \) MPas.

The evolution of \( C_{11}^v \) is shown in dashed line on Fig. 4.

The computation of the PK2 stress tensor needs further development: the shift in the hydrostatic pressure space \( p_0 \) is obtained by imposing the initial condition (stress free for no strain) \( S_{11} = S_{22} = S_{33} = 0 \) for \( \lambda = C_{11}^v = 1 \). Therefore, \( p_0 = 2(c_{10}^v + 2c_{01}^v) \).

The \( p \) Lagrange multiplier is eliminated by applying the uniaxial condition: \( S_{22} = S_{33} = 0 \) whatever \( \lambda \) and \( C_{11}^v \).

The \( S_{11} \) relaxation is plotted in solid line in Fig. 4 (right y-axis) whereas both \( S_{22} \) and \( S_{33} \) are identically zero.

3.6.2. Worked example 2: uniaxial tension/tension cyclic test

The deformation gradient tensor and the material coefficients are the same as in the worked example 1. Only the loading condition is changed: \( \lambda = 2 - \cos(\pi t) \). The results are summarized in Fig. 5. On the left diagram, the evolution of \( C_{11}^v \) (solid

![Fig. 4. Evolution of \( C_{11}^v \) and the stress relaxation.](image-url)
line) obtained by imposing \( \lambda \) (dashed line) is plotted. The right diagram illustrates the PK2 stress versus \( \lambda \). Note that the calculation is run notwithstanding the compressive stress. We can reasonably conclude that the response of the constitutive model is relevant.

4. Mullins’ effect: discontinuous damage model

The so-called Mullins’ effect is a strain-induced stress softening of filled rubber material. Many workers have brought their contributions on the understanding of this phenomenon (Mullins and Tobin, 1957; Govindjee and Simo, 1991; Miehe, 1995; Lion, 1996, etc).

Experimentally, it is observed that the first loading path (in a cyclic stress–strain curve) is different from the unloading path. This latter will then be common to the following cycles (loading/unloading) provided that the applied strain does not overrun the maximum strain reached in the history of deformation. The micromechanisms responsible for this effect are attributed by many authors (Bueche, 1967; Govindjee and Simo, 1991) to the composite material which consists of a rubber matrix and the fillers. According to these authors, there is relative motion of the filler and the rubber matrix, which may lead to a local separation of the filler and the matrix. For the material of interest, microscopic examinations have been carried out on a strip stretched in situ using the scanning electron microscope (SEM). Fig. 6 clearly shows that for a very low value of \( \lambda \) stretch, disruption of a silica agglomerate occurs. This can be related to surface creation during the early stage of the stretching.

Following this idea, we will describe the Mullins’ effect as a continuum damage model driven by the strain maximum value. Note that many other researchers have modeled Mullins’ effect in this way (Miehe, 1995; Lion, 1996). Let \( D \) \((0 \leq D < 1)\) be the scalar internal variable which describes an isotropic damage effect: \( D \) is a relative measure for the damaged area (e.g. Kachanov, 1986; Lemaitre and Chaboche, 1988;
The nominal Helmholtz free energy is assumed to be given by the constitutive expression: \( W = (1 - D)W_0 \), where \( W_0 \) is the effective free energy with respect to the undamaged material. In the following, \( W_0 \) is the free energy accounting for the viscoelasticity effect. The multiplicative decomposition of the nominal free energy allows to separate analysis of the damage term \((1 - D)\) and the aforementioned visco-hyperelasticity study. In particular, Eq. (1) can be extended to:

\[
S - 2 \frac{\partial W}{\partial \dot{\varepsilon}} : \dot{\varepsilon} - 2 \frac{\partial W}{\partial \dot{A}_i} \dot{A}_i - 2 \frac{\partial W}{\partial \dot{D}} \dot{D} \geq 0
\]  

(38)

Let \( f \) be the thermodynamic force which drives the damage evolution:

\[
f = \frac{\partial W}{\partial D} = \frac{\partial[(1 - D)W_0]}{\partial D} = -W_0
\]  

(39)

To determine the evolution of damage, a surface \( s = 0 \) is introduced.

\[
s(f, Q) = f - Q
\]  

(40)

Following Miehe (1995), we assume that damage is governed by the variable \( Q(D, t) = \text{Max}_{s \in [0, t]} f(s) \). \( Q \) is the maximum thermodynamic force which has been achieved in the history interval \([0, t]\). Thus, the evolution of \( Q \) becomes:

\[
\dot{Q} = \begin{cases} 
\dot{f} & \text{for } f - Q = 0 \text{ and } \dot{f} > 0 \\
0 & \text{otherwise}
\end{cases}
\]  

(41)

with the initial condition \( Q(D, t = 0) = 0 \). This underlines the discontinuous character of this damage: there is no damage accumulation if the thermodynamic force \( f \) lies inside a damage domain bounded by \( s \) (i.e. \( f - Q < 0 \)).

A suitable form of \( Q(D) \) is the following (Robisson, 2000):

\[
Q(D) = -\eta \ln \left( \frac{D}{D_\infty} \right)
\]  

(42)
$D_\infty$ and $\eta$ are material coefficients whose physical meaning will be developed further. Since

$$\dot{Q} = \frac{\partial Q}{\partial D} \dot{D}$$

It follows that:

$$\int_0^Q \frac{dQ}{\eta} = \int_0^D \frac{dD}{D_\infty - D}$$

After integration, the damage evolution proposed by Miehe (1995) is retrieved:

$$D = D_\infty \left(1 - e^{-\frac{Q}{\eta}}\right)$$

Remind that $Q$ is the maximum thermodynamic force which has been achieved with the history interval $[0, t]$. Moreover, the significance of $D_\infty$ and $\eta$ material coefficients is clear: they represent respectively the maximum possible damage and the damage saturation parameter.

In the following, we assume that:

$$Q = \lambda^I_{\max} - 1$$

where $\lambda^I_{\max}$ is the maximum largest principal stretch: superscript $I$ denotes that it is the largest of the three principal stretches, whereas the subscript max stands for the maximum value reached by this $\lambda^I$ stretch during the history interval $[0, t]$. The damage evolution [Eq. (44)] becomes:

$$D = D_\infty \left(1 - e^{-\frac{\lambda^I_{\max} - 1}{\eta}}\right)$$

5. Finite element analysis

The above constitutive model has been implemented in the home made FE code Z-set (Besson and Foerch, 1997). Mixed (or hybrid) elements are utilized with the total Lagrangian formulation. Mullins’ effect (discontinuous damage model) is accounted for. The verification is made by comparing the experimental cyclic response of the silica filled rubber with the finite element simulation. Finally, a validation of both the model and the material coefficients is checked out by numerical simulations of experimental tests on more complex specimen geometries.

5.1. Optimization of the material parameters

The Z-set FE code is provided with optimization routines involving various optimization methodologies. All the constitutive equations coupled with the discontinuous
damage concepts require the determination of 8 material coefficients. Some of these unknowns are subjected to constraints such as: $D_\infty < 1$ or $c_{ij}^\tau > 0$. These latter — sufficient but not necessary — conditions ensure the Drucker stability consisting in maintaining positive the energy due to an infinitesimal change in the strain. When some Rivlin coefficients are negative, it may induce instability in the stress–strain curve. From our point of view, the most efficient optimization method for handling 8 unknowns with some constraints is the evolutionary algorithm (Besson et al., 1998).

The optimization strategy consists in:

- uncoupling the phenomena so that the hyperelastic, damage and viscoelastic coefficients be identified individually using appropriate experimental data;
- doing an additional optimization on these pre-identified coefficients by choosing experimental data where all phenomena are intermixed like uniaxial cyclic tension to tension tests.

The set of material coefficients used in the worked examples comes from the optimization procedure. Only $D_\infty$ and $\eta$ damage parameters have not been presented. The whole set of coefficients are:

- The Rivlin strain energy for the first spring
  \[ W_c = c_{10}^e (I_1^c - 3) + c_{01}^e (I_2^c - 3) + c_{20}^e (I_2^e - 3)^2 \]
  \( c_{10}^e (\text{MPa}) = 3.565 \)
  \( c_{01}^e (\text{MPa}) = 1.305 \)
  \( c_{20}^e (\text{MPa}) = 0.839 \)

- The Mooney Rivlin strain energy for the second spring
  \[ W_v = c_{10}^v (I_1^v - 3) + c_{01}^v (I_2^v - 3) \]
  \( c_{10}^v (\text{MPa}) = 3.470 \)
  \( c_{01}^v (\text{MPa}) = 1.866 \)

- The “viscosity” coefficient $f_v = 47$ MPas
- The discontinuous damage model parameters are $D_\infty = 0.718$ and $\eta = 0.206$.

5.2. Volume element simulation

In this subsection, we need to get back to Fig. 1, where the experimental curve shows the three aspects of the mechanical behavior at once. The numerical simulation is performed on one axisymmetric hybrid element of unit length, with full integration (9 integration points) and 4 pressure nodes. Fig. 1 shows the comparison between finite element results (dashed line) and experimental data (solid line). The two curves are in rather good agreement. However, it is noticeable that neither the first loading path nor the area of the hysteresis loop do match the experimental data. In fact, the main goal of the study is the fatigue lifetime of the prescribed material and it essentially requires to model the cyclically stabilized amplitude of the stress/
strain and the permanent elongation measured at zero stress level. It is clear that these two quantities are correctly modeled in Fig. 1.

5.3. Validation on NS40 specimen, 2D simulation

This subsection aims at validating the aforementioned optimized set of material parameters. The experimental data come from the NS40 specimen (Fig. 2, left) subjected to cyclic tension to compression loading. The notch radius sets an inhomogeneous stress/strain state in the vicinity of the notch root. Moreover, it is possible to impose a compression part in the loading (no buckling). The FE meshing involves only a quarter of the NS40 specimen due to symmetry. Two-dimensional axisymmetric mixed elements are utilized, with reduced integration. The material parameters are the same as in the previous subsection, including the damage model. Fig. 7 shows that the FE results (dashed line) match very well experimental data (solid line), even in the compressive area of the diagram. This kind of test was not used in the material parameters optimization process database. Accordingly, the plot of Fig. 7 validates the constitutive equations and the optimized material parameters.

5.4. Validation on NS2 specimen, 3D simulation

The extension to the 3D case of the simulation is presented in this subsection. For this purpose, the NS2 specimen (Fig. 2, right) is meshed with 3D elements with reduced integration. The meshing consists of only half of the specimen. The loading condition is a cyclic torsion between 10° and 40°, involving a shear mode mainly. In addition, a stress/strain gradient is encountered inside the specimen.

Fig. 8 (left) shows the comparison between FE results (dashed line) and experimental data (solid line). The FE simulation overestimates the stiffness of the specimen. This is due to

---

Fig. 7. Load vs. displacement experimental and simulated curves under cyclic loading.
• the lack in experimental results of shear tests in the optimization procedure, so the coefficient set does not correctly reflect the shear behavior of the material;
• the lack in some relaxation times in the model that uses a unique coefficient $f_v$ to take the viscoelasticity into account, whereas in a previous work that utilised the hereditary integral (Robisson, 2000), the Prony series were provided with 5 terms.

The last point raises the problem of extension of the model in order to account for more than one relaxation time. This can be done either by considering the Zener model and adding the required number of spring-dashpots in parallel, or by keeping the Poynting–Thomson model and rendering the relation depicted in Eq. (14) nonlinear. Nevertheless, this last comparison indicates that a fully 3D computation using such constitutive equations can be completed.

The FE code allows to compute the stress/strain tensors at any location (integration point) of a “structure”. As far as the global (measurable) parameters such as the load vs. displacement or the torque vs. angle curves are correctly simulated by the FE code, we can assume that the local parameters (stress/strain tensors) are satisfactorily calculated. As an illustration, Fig. 8 (right) displays both the NS2 specimen meshing in 3D and the contour map of the damage parameter $D$ (denoted by dmg here). Any local parameter ($\sigma_{ij}$, $E_{ij}$, $C_{ij}$, ...) can be accessed thanks to these FE results. For the fatigue lifetime analysis, this is very useful because every local parameter can be analyzed in order to determine which one is relevant to describe the crack initiation (Robisson, 2000).

6. Closure

The silica-filled rubber material presented in this paper exhibits nonlinear elasticity, nonlinear rate dependence and Mullins’ effect.

Fig. 8. Torque vs. angle (experimental and simulated curves) under cyclic loading and damage parameters contour map on the deformed mesh of NS2 specimen.
The nonlinear elasticity is accounted for by using the Rivlin’s deformation energy model.

The viscoelasticity theory under the finite deformation aspect relies on the tensorial ISV proposed by Sidoroff (1975a). In addition, the evolution law is treated by defining a dissipation pseudo-potential (Germain, 1986). The multiplicative split of the deformation gradient—and by the way, the intermediate configuration concept—is assumed. As opposed to Lion (1996, 1997) or Reese and Govindjee (1998), we consider the Poynting–Thomson rheological model that is based upon the instantaneous (infinitely high strain rate) elasticity.

Mullins’ effect is analyzed with the help of a discontinuous damage concept (Miehe, 1995; Govindjee and Simo, 1991). This part is separated from the viscoelasticity approach.

As a result, the constitutive equations are established and a calculation method is suggested. Analytical simulations are presented followed by FE analyses with 2D and 3D geometries. The constitutive equations with their optimized set of parameters are validated by comparing the simulated results with the experimental data. Accordingly, the confidence on the computation of every local parameter is rather high. Using this method the fatigue lifetime of the prescribed silica—filled rubber material can then be addressed by plotting a relevant local parameter against the number of cycles to crack initiation.

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Appendix

In this Appendix, we describe in detail the calculations of the first derivative of invariants with respect to the right Cauchy–Green tensor. Einstein’s notation is used and the Kronecker’s symbol is defined as follows:

\[
\delta_{ij} = \begin{cases} 
1 & \text{if } i = j \\
0 & \text{otherwise}
\end{cases}
\]

Calculation of \(\partial I_1^e / \partial C\)

By definition of the first invariant of the right Cauchy–Green tensor:

\[
I_1^e = C_{ii}^e = F_{ik}^v C_k F_{li}^{v-1} = F_{li}^{v-1} F_{li}^{v-1} C_{kl} = C_{lk}^{-1} C_{kl}
\]

Note that:

\[
I_1^e = C^{v-1} : C
\]
Since $\mathbf{C}^{r-1}$ is symmetric $\mathbf{C}_{nn}^{r-1} = \mathbf{C}_{nn}^{r-1}$, it follows that:

$$\frac{\partial I_2^e}{\partial \mathbf{C}} = \mathbf{C}^{r-1}$$

This result is used in [Eq. (19)].

**Calculation of $\partial I_2^e / \partial \mathbf{C}$**

By definition of the second invariant of the right Cauchy–Green tensor:

$$I_2^e = \frac{1}{2} \left( I_{2e}^2 - \mathbf{C}_{yi}^{r} \mathbf{C}_{ji}^{r} \right)$$

So:

$$\frac{\partial I_2^e}{\partial \mathbf{C}} = I_{2e}^{r} \frac{\partial I_{2e}^e}{\partial \mathbf{C}} - \frac{1}{2} \frac{\partial \left( \mathbf{C}_{yi}^{r} \mathbf{C}_{ji}^{r} \right)}{\partial \mathbf{C}}$$

[Eq. (48)] allows to write:

$$\frac{\partial I_2^e}{\partial \mathbf{C}} = I_{2e}^{r} \mathbf{C}^{r-1} - \frac{1}{2} \frac{\partial \left( \mathbf{C}_{yi}^{r} \mathbf{C}_{ji}^{r} \right)}{\partial \mathbf{C}}$$

Let us proceed to $\mathbf{C}_{yi}^{r} \mathbf{C}_{ji}^{r}$ arrangement:

$$\mathbf{C}_{yi}^{r} \mathbf{C}_{ji}^{r} = \mathbf{F}_{ik}^{r-1} \mathbf{C}_{kl} \mathbf{F}_{lj}^{r-1} \mathbf{F}_{kr}^{r-1} \mathbf{F}_{rs} \mathbf{F}_{ii}^{r-1} = \left( \mathbf{F}_{ii}^{r-1} \mathbf{F}_{jl}^{r-1} \mathbf{F}_{jr}^{r-1} \right) \mathbf{C}_{kl} \mathbf{C}_{rs}$$

$$= \mathbf{C}_{sk}^{r-1} \mathbf{C}_{ij}^{r-1} \mathbf{C}_{sr} = \left( \mathbf{C}^{r-1} \mathbf{C}^{r-1} \mathbf{C}^{r-1} \right)_{ls} \left( \mathbf{C}^{r-1} \mathbf{C}^{r-1} \mathbf{C}^{r-1} \right)_{ls} = \left( \mathbf{C}^{r-1} \mathbf{C}^{r-1} \mathbf{C}^{r-1} \right)$$

Hence,

$$\mathbf{C}_{yi}^{r} \mathbf{C}_{ji}^{r} = \left( \mathbf{C}^{r-1} \mathbf{C}^{r-1} \mathbf{C}^{r-1} \mathbf{C}^{r-1} \right)$$

Derivating [Eq. (49)] with respect to $\mathbf{C}$,

$$\left( \frac{\partial \left( \mathbf{C}^{r-1} \mathbf{C}^{r-1} \mathbf{C}^{r-1} \mathbf{C}^{r-1} \right)}{\partial \mathbf{C}} \right)_m = \left( \frac{\partial \left( \mathbf{C}^{r-1} \mathbf{C}^{r-1} \mathbf{C}^{r-1} \mathbf{C}^{r-1} \right)}{\partial \mathbf{C}} \right)_m = \left( \frac{\partial \left( \mathbf{C}^{r-1} \mathbf{C}^{r-1} \mathbf{C}^{r-1} \mathbf{C}^{r-1} \right)}{\partial \mathbf{C}} \right)_m$$

$$= \mathbf{C}_{il}^{r-1} \mathbf{C}_{jk}^{r-1} \frac{\partial \mathbf{C}_{lk}}{\partial \mathbf{C}_{mn}} \mathbf{C}_{ij}^{r-1} + \mathbf{C}_{il}^{r-1} \mathbf{C}_{jk}^{r-1} \frac{\partial \mathbf{C}_{lk}}{\partial \mathbf{C}_{mn}} \mathbf{C}_{ij}^{r-1} = \mathbf{C}_{ij}^{r-1} \mathbf{C}_{ij}^{r-1} \mathbf{C}_{ij}^{r-1} \mathbf{C}_{ij}^{r-1} \mathbf{C}_{ij}^{r-1} \mathbf{C}_{ij}^{r-1} \mathbf{C}_{ij}^{r-1} \mathbf{C}_{ij}^{r-1} \mathbf{C}_{ij}^{r-1} \mathbf{C}_{ij}^{r-1} \mathbf{C}_{ij}^{r-1} \mathbf{C}_{ij}^{r-1}$$

$$= \mathbf{C}_{im}^{r-1} \mathbf{C}_{jn}^{r-1} \mathbf{C}_{ij}^{r-1} + \mathbf{C}_{im}^{r-1} \mathbf{C}_{jn}^{r-1} \mathbf{C}_{ij}^{r-1}$$
because \( \mathbf{C}^{v-1} \) is symmetric;

\[
\frac{\partial \left( \mathbf{C}^{v-1} \mathbf{C} \right)}{\partial \mathbf{C}} : \left( \mathbf{C} \mathbf{C}^{v-1} \right) = 2\mathbf{C}^{v-1} \mathbf{C} \mathbf{C}^{v-1}
\]

(50)

Finally:

\[
\frac{\partial I^e_2}{\partial \mathbf{C}} = I_1^e \mathbf{C}^{v-1} - \mathbf{C}^{v-1} \mathbf{C} \mathbf{C}^{v-1}
\]

(51)

This result is used in Eq. (20).

**Calculation of \( \frac{\partial I^e_2}{\partial \mathbf{C}^v} \)**

Remember that Eq. (47) \( I^e_2 = \mathbf{C}^{v-1} : \mathbf{C} \)

Hence,

\[
\left( \frac{\partial I^e_2}{\partial \mathbf{C}} \right)_{mn} = \frac{\partial I^e_1}{\partial \mathbf{C}^{v-1}} = \frac{\partial \mathbf{C}^{v-1}_{ik}}{\partial \mathbf{C}^{v}_{mn}} \mathbf{C}^{kl}
\]

Starting with: \( \mathbf{C}^{v-1} \mathbf{C}^v = \mathbf{I} \) or \( \mathbf{C}_{il}^{v-1} = \mathbf{C}_{ln}^v = \delta_{in} \)

one can write that:

\[
\frac{\partial \left( \mathbf{C}^v_{ij} \mathbf{C}^{v-1}_{jk} \right)}{\partial \mathbf{C}^v_{mn}} = \delta_{im}\delta_{jn} \mathbf{C}^{v-1}_{jk} + \mathbf{C}^v_{ij} \frac{\partial \mathbf{C}^{v-1}_{jk}}{\partial \mathbf{C}^v_{mn}} = 0
\]

Let us multiply the last equation by \( \mathbf{C}^v_{li} \):

\[
\delta_{im}\delta_{jn} \mathbf{C}_{li}^{v-1} \mathbf{C}^{v-1}_{jk} \mathbf{C}^v_{ij} \frac{\partial \mathbf{C}^{v-1}_{jk}}{\partial \mathbf{C}^v_{mn}} = 0
\]

\[
\mathbf{C}^{v-1}_{lm} \mathbf{C}^{v-1}_{nk} + \delta_{ij} \frac{\partial \mathbf{C}^{v-1}_{jk}}{\partial \mathbf{C}^v_{mn}} = 0
\]

Therefore:

\[
\frac{\partial \mathbf{C}^{v-1}_{ik}}{\partial \mathbf{C}^v_{mn}} = -\mathbf{C}^{v-1}_{lm} \mathbf{C}^{v-1}_{nk}
\]

(52)
Then,
\[
\left( \frac{\partial I^e}{\partial C^v} \right)_{mn} = \frac{\partial C^{-1}_{jk}}{\partial C^v_{mn}} C_{kl} = -C^{-1}_{lm} C^{-1}_{nk} C_{kl}
\]

Since $C$ and $C^{v-1}$ are symmetric: $C^{v-1}_{lm} = C^{-1}_{nk} C_{kl} = C^{v-1}_{ml} C^{v-1}_{lk}$

Finally:
\[
\frac{\partial I^e_i}{\partial C^v} = C^{v-1} C^{v-1}
\]

(53)

This result is used in Eq. (22).

**Calculation of $\frac{\partial I^e_2}{\partial C^v}$**

By definition,
\[
I^e_2 = \frac{1}{2} \left( I^e_1 - C^{e}_{ij} C^{e}_{ji} \right)
\]

\[
\frac{\partial I^e_2}{\partial C^v} = I^e_1 \frac{\partial I^e_1}{\partial C^v} - \frac{1}{2} \frac{\partial (C^{e}_{ij} C^{e}_{ji})}{\partial C^v} = -I^e_1 C^{v-1} C^{v-1} - \frac{1}{2} \frac{\partial (C^{e}_{ij} C^{e}_{ji})}{\partial C^v}
\]

Eq. (49) reads:
\[
C^{e}_{ij} C^{e}_{ji} = \left( C^{v-1} C \right) : \left( C C^{v-1} \right)
\]

and Eq. (50) reads:
\[
\frac{\partial \left( C^{v-1} C : C C^{v-1} \right)}{\partial C^{v-1}} = 2 C^{v-1} C^{v-1}
\]

The latter equation [Eq. (50)] allows us to write that:
\[
\frac{\partial \left( C^{v-1} C : C C^{v-1} \right)}{\partial C^{v-1}} = 2 C C^{v-1}
\]

It follows that:
\[
\frac{\partial \left( C^{e}_{ij} C^{e}_{ji} \right)}{\partial C^v} = \frac{\partial \left( C^{v-1} C : C C^{v-1} \right)}{\partial C^v}
\]

\[
\left( \frac{\partial \left( C^{v-1} C : C C^{v-1} \right)}{\partial C^v} \right)_{mn} = \frac{\partial \left( C^{v-1} C : C C^{v-1} \right)}{\partial C^v_{mn}} = \frac{\partial \left( C^{v-1} C : C C^{v-1} \right)}{\partial C^{v-1}_{kl}} \frac{\partial C^{v-1}_{kl}}{\partial C^v_{mn}}
\]
Moreover Eq. (52),

\[
\frac{\partial C_{v}^{-1}}{\partial C_{mn}} = -C_{km}^{-1} C_{ln}^{-1}
\]

Then:

\[
\left( \frac{\partial \left( C_{v}^{-1} C : C C_{v}^{-1} \right)}{\partial C_{v}} \right)_{nn} = \left( 2C C_{v}^{-1} C \right)_{kl} \left( C_{km}^{-1} C_{ln}^{-1} \right) = -2C_{mk}^{-1} \left( C C_{v}^{-1} C \right)_{kl} C_{ln}^{-1}
\]

Hence:

\[
\frac{\partial \left( C_{v}^{-1} C : C C_{v}^{-1} \right)}{\partial C_{v}} = -2C_{v}^{-1} \left( C C_{v}^{-1} C \right) C_{v}^{-1}
\]

And finally:

\[
\frac{\partial I_{e}}{\partial C_{v}} = -I_{e1} C_{v}^{-1} C C_{v}^{-1} + C_{v}^{-1} \left( C C_{v}^{-1} C \right) C_{v}^{-1}
\]

Eq. (54) is identical to Eq. (23).

References


