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Modeling of nonlinear viscoelasticity at large deformations

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Abstract A constitutive model of finite strain viscoelasticity, based on the multiplicative decomposition of the deformation gradient tensor into elastic and inelastic parts, is presented. The nonlinear response of rubbers, manifested by the rate effect, cycling loading and stress relaxation tests was captured through the introduction of two internal variables, namely the constitutive spin and the back stress tensor. These parameters, widely used in plasticity, are applied in this work to model the nonlinear viscoelastic behaviour of rubbers. The experimental results, obtained elsewhere, related with shear deformation in monotonic and cyclic loading, as well as stress-relaxation, were simulated with a good accuracy.

Introduction

The theoretical study of the mechanics of nonlinear materials with memory was first performed by Green and Rivlin [1], Coleman and Noll [2], Noll [3], and Pipkin [4]. Green and Rivlin have examined materials where the stresses were functionally dependent on the deformation gradient history, in terms of multiple integration. Valanis and Landel [5] have proposed a constitutive equation at large deformations for a filled rubber. This equation contains three relaxation functions proportional to the same function of time and independent of the deformation.

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Finite strain viscoelasticity was also studied by Flowers and Lianis [6] where theoretical expressions for uniaxial and equal homogeneous biaxial single-step and double-step relaxation were developed.

The nonlinear viscoelastic response of filled rubbers, revealed either by strain rate dependence or stress relaxation behaviour, remains still one of the most intricate tasks for finite viscoelasticity. The effect of strain rate on the mechanical response of filled rubber was studied earlier by Mason [7] and Dannis [8], when the tensile strength of rubber was found to increase with increasing strain rate. Apart from this, when an elastomer is subjected to constant strain, and the corresponding stress response is recorded, it was found by Gent [9] that the stress relaxes significantly during the first 2 s. Therefore, the stress relaxation time history during the first 6 s of the tests remained unknown. Later, this short coming has been overcome, and in subsequent studies of Lion [10], Bergstrom and Boyce [11, 12], more information about stress relaxation history of rubbers under uniaxial tension and compression has been reported.

Hereafter, creep, relaxation and rate dependent behaviour of polymers under tension was investigated by Khan and Zhang [13], Khan and Lopez-Pamies [14] and Krempl and Khan [15]. Moreover, strain rate sensitivity, multiple creep and recovery behaviour of polyphenylene oxide, studied experimentally by Khan [16], were modeled by Colak [17], in terms of a viscoplasticity theory based on overstress. Nonlinear rate sensitivity, nonlinear unloading, creep and recovery data could be reproduced using the theory of overstress. Within this frame, in a work by Naghidabadi et al. [18] a finite deformation constitutive model for rigid plastic hardening materials based on the logarithmic strain tensor is introduced. The flow rule of this constitutive model relates the corotational rate of the logarithmic strain tensor to the difference of the deviatoric Cauchy stress and the back stress tensor. The evolution equation for the kinematic hardening of this model relates the corotational rate of the back stress tensor to the corotational rate of the logarithmic strain tensors. In a work by Kafka et al. [19], modeling of creep and relaxation is presented in good agreement with experimental results. In this approach, notions like back stress, effective stress, overstress and equilibrium stress are given clear physical meaning by confronting their use with the use of internal stresses, represented on the mesoscale as tensorial internal variables.

Vulcanized rubbers are related with a wide range of engineering applications, such as tires, engine mounts, tunnel linings or buildings protection from earthquakes. Rubber bearings for base isolation devices are usually of cubic and cylindrical shape and are mainly subjected to a combination of compression and shear deformation.

When the structure is subjected to large cyclic loading, arising from earthquakes the delivered energy may be absorbed through the rubber's hysteresis properties. Therefore, high damping rubbers (HDR) are synthesized with a large number of additives such as carbon black, silica and oils. The quasistatic cyclic behaviour of HDR under shear has been reported by Yoshida et al. [20]. Moreover, Amin [21] and Amin et al. [22], examined the nonlinear rate-independent monotonic response of the equilibrium stress of HDR, in terms of compression and shear. In their study, an important hyperelasticity relation, a procedure for the identification of material parameters and the application of the model in a general purpose finite element code were proposed to simulate rate independent response. However, it was shown from these studies, that there is a significant rate-dependence effect in HDR.

Therefore, due to the above-mentioned reasons, there is a need, for the estimation of the performance of rubber bearings and consequently their optimum design.

In a recent work by Amin et al. [22], the rate dependent behaviour of filled natural rubber (NR) and HDR has been studied in compression and shear. A constitutive model of finite strain viscoelasticity, based on the multiplicative decomposition of the deformation gradient tensor into elastic and inelastic parts is proposed. The total stress is decomposed into an equilibrium stress and a viscosity-induced overstress. An evolution equation in terms of power laws was proposed to express the effects of internal variables on viscosity phenomena.

Their analysis is based on a work by Huber et al. [23], where an analogous decomposition in terms of two types of spring-dashpot models is presented. In their approach, the second law of thermodynamics was satisfied, and the concept of the so-called Mandel stress-tensor was applied. More specifically, from the requirement of the thermodynamic consistency, a flow rule for the inelastic strain rate in the so-called intermediate configuration was proved to apply, and was expressed in terms of an overstress, formed by Mandel stress tensors.

In a series of works by Boyce et al. [24], macroscopic mechanical behaviour was connected with molecular mechanisms of deformation resistance, through three-dimensional constitutive models. The rate-, temperature-, and pressure-dependent finite-strain deformation of thermoplastic materials was described with a model composed of a linear spring in series with a viscoplastic dashpot to express the intermolecular resistance, in parallel with a nonlinear Langevin spring, representing the entropic hardening. The corresponding constitutive laws connect the rates of shape change (plastic stretching) with the stress in the deforming material. The direction tensor is taken to be coaxial with the deviatoric stresses acting on the intermolecular network.

In the present analysis, it is assumed that a material's finite deformation viscoelasticity arises from an intermolecular resistance, related with both an elastic constituent and a nonlinear fluid, as well as from an entropic resistance. The last one is modeled with a non-Gaussian type constitutive equation for elastomers. Moreover, the evolution of the viscoelastic component is mainly determined by an internal variable, the so-called back stress tensor $\boldsymbol{\alpha}$. The concept of constitutive spin introduced by Dafalias [25, 26] was also used to express the objective rates of the constitutive equation of hypoelasticity. Both variables, back stress and constitutive spin were applied to the constitutive laws of the symmetric and antisymmetric part of the inelastic velocity gradient tensor. The main features of nonlinear viscoelastic response of rubbers, such as rate effect on monotonic and cyclic loading, as well as stress relaxation behaviour in terms of shear deformation were captured with a very satisfactory approximation. Corresponding experimental results were available from a recent work by Amin et al. [22].

Kinematics

In the present work, finite deformation viscoelasticity will be treated within the framework of the multiplicative decomposition of the deformation gradient tensor $\mathbf{F}$ into an elastic $\mathbf{F}^{\mathbf{e}}$ and inelastic (viscoelastic) part $\mathbf{F}^{\mathbf{i}}$, in a similar way to the corresponding multiplicative decomposition of $\mathbf{F}$ in plasticity (see [27] and [28]).

According to this decomposition, expressed in Eq. 1, part $\mathbf{F}^{\mathbf{i}}$ introduces an intermediate equilibrium configuration, which results from the current state by fast unloading, or from the original configuration by inelastic deformation.

$$\mathbf{F} = \mathbf{F}^{\mathbf{e}}\mathbf{F}^{\mathbf{i}}. \tag{1}$$

Subsequently, the elastic deformation gradient tensor $\mathbf{F}^{\mathbf{e}}$ maps the material point from the relaxed to the current configuration, which is obtained from the relaxed by purely elastic deformation and rotation. The relaxed configuration is arbitrarily defined, since an arbitrary rigid rotation can be superimposed on it and leave it unstressed.

The velocity gradient tensor $\mathbf{L}$ in the current configuration is defined by:

$$\mathbf{L} = \dot{\mathbf{F}}\mathbf{F}^{-1} = \mathbf{D} + \mathbf{W} \tag{2}$$

and at the unstressed configuration:

$$\dot{\mathbf{F}}^{\mathbf{i}}\mathbf{F}^{\mathbf{i}^{-1}} = \left(\dot{\mathbf{F}}^{\mathbf{i}}\mathbf{F}^{\mathbf{i}^{-1}}\right)_{\mathrm{s}} + \left(\dot{\mathbf{F}}^{\mathbf{i}}\mathbf{F}^{\mathbf{i}^{-1}}\right)_{\mathrm{a}} = \mathbf{D}_{\mathbf{0}}^{\mathbf{i}} + \left(\dot{\mathbf{F}}^{\mathbf{i}}\mathbf{F}^{\mathbf{i}^{-1}}\right)_{\mathrm{a}} \tag{3}$$

where $\mathbf{D},\mathbf{W}$ are the rate deformation tensor and material spin tensors correspondingly at the current configuration. Subscripts s, a denote the symmetric and antisymmetric part of a tensor, and $\mathbf{D}_{\mathbf{o}}^{\mathbf{i}}, (\dot{\mathbf{F}}^{\mathbf{i}}\mathbf{F}^{\mathbf{i}^{-1}})_{\mathrm{a}}$ are the inelastic rate of deformation tensor and inelastic material spin tensor at the relaxed configuration. It is important to mention here again, what is emphasized by Dafalias [29], i.e., the arbitrariness of the term $(\dot{\mathbf{F}}^{\mathbf{i}}\mathbf{F}^{\mathbf{i}^{-1}})_{\mathrm{a}}$, which is related with the arbitrariness of the intermediate configuration.

To overcome this problem, Mandel [30] proposed a triad of orthonormal vectors that represent the material substructure and follow different kinematics than the continuum. The kinematic quantities $\mathbf{F}^{\mathbf{e}}$ and $\mathbf{F}^{\mathbf{i}}$ are then defined in terms of the corotational rates as follows:

$$\begin{aligned} \overset{\mathrm{o}}{\mathbf{F}}{}^{\mathrm{e}} &= \dot{\mathbf{F}}^{\mathbf{e}} + \mathbf{F}^{\mathbf{e}}\boldsymbol{\omega} \\ \overset{\mathrm{o}}{\mathbf{F}}{}^{\mathrm{i}} &= \dot{\mathbf{F}}^{\mathbf{i}} - \boldsymbol{\omega}\mathbf{F}^{\mathbf{i}} \end{aligned} \tag{4}$$

where $\boldsymbol{\omega}$ is the spin of the substructure related with the rotation of the triad vestors. This is in accordance with the common notion in elasticity theory, that elastic deformation is directly connected to the deformation of the substructure, while this cannot be generalized in anelasticity. In the elastic–plastic response, a decoupling deformation process between continuum and substructure is required.

Following this consideration and taking into account, Eq. (4), the kinematics in the current configuration is given by:

$$\mathbf{D} = \mathbf{D}^{\mathbf{e}} + \mathbf{D}^{\mathbf{i}} \tag{5}$$

where $\mathbf{D}^{\mathbf{e}}$ is the elastic part of the rate deformation tensor, and $\mathbf{D}^{\mathbf{i}}$ the inelastic part of it. All these quantities are now invariant upon superposed rigid body rotation.

The corresponding relations are as follows:

$$\mathbf{D}^{\mathbf{e}} = \left(\overset{\mathbf{0}}{\mathbf{F}^{\mathbf{e}}}\mathbf{F}^{\mathbf{e}^{-1}}\right)_{\mathrm{s}} \tag{6}$$

$$\mathbf{D}^{\mathbf{i}} = \left(\mathbf{F}^{\mathbf{e}}\left[\mathbf{D}_{\mathbf{0}}^{\mathbf{i}} + \left(\overset{\mathbf{0}}{\mathbf{F}^{\mathbf{i}}}\mathbf{F}^{\mathbf{i}^{-1}}\right)_{\mathrm{a}}\right]\mathbf{F}^{\mathbf{e}^{-1}}\right)_{\mathrm{s}} \tag{7}$$

and

$$\mathbf{D}_{\mathbf{0}}^{\mathbf{i}} = \left[\overset{\mathbf{0}}{\mathbf{F}^{\mathbf{i}}}\mathbf{F}^{\mathbf{i}^{-1}}\right]_{\mathrm{s}} \tag{8}$$

With respect to the corotational rates for the kinematical quantities, it follows also Dafalias [26] that:

$$\left(\dot{\mathbf{F}}^{\mathbf{i}}\mathbf{F}^{\mathbf{i}^{-1}}\right)_{\mathrm{a}} = \boldsymbol{\omega} + \left(\overset{\mathbf{0}}{\mathbf{F}^{\mathbf{i}}}\mathbf{F}^{\mathbf{i}^{-1}}\right)_{\mathrm{a}} = \boldsymbol{\omega} + \mathbf{W}_{\mathbf{0}}^{\mathbf{i}} \tag{9}$$

where $\mathbf{W}_{\mathbf{0}}^{\mathbf{i}}$ is the inelastic spin at the intermediate configuration material and $\boldsymbol{\omega}$ is the substructural spin. Through this relation, which is analogous to Eq. 5 obtained from the additive decomposition of the deformation rate tensor, the arbitrariness of $(\dot{\mathbf{F}}^{\mathbf{i}}\mathbf{F}^{\mathbf{i}^{-1}})_{\mathrm{a}}$ can be overcome with a proper value of $\boldsymbol{\omega}$. Referring to Eq. 9, Mandel [30] has considered $\boldsymbol{\omega}$ to be the rigid body spin of the director triad, common for all tensorial internal variables. Later Dafalias [29] generalized this description, introducing different $\boldsymbol{\omega}$'s associated with each internal variable, and abandoned the notion of director triad as unnecessary. The decomposition as expressed by Eqs. 5 and 9 calls for constitutive laws for $\mathbf{D}^{\mathbf{e}}, \mathbf{D}_{\mathbf{0}}^{\mathbf{i}}, \mathbf{W}_{\mathbf{0}}^{\mathbf{i}}$ and $\boldsymbol{\omega}$.

For the implementation of our analysis to the nonlinear, large deformation response, an internal variable is introduced, in terms of a deviatoric back-stress tensor $\boldsymbol{\alpha}$.

The necessity of constitutive relations for $\mathbf{D}_{\mathbf{0}}^{\mathbf{i}}, \mathbf{W}_{\mathbf{0}}^{\mathbf{i}}$ is in strong connection with this internal variable. To provide a definite expression for $\mathbf{D}^{\mathbf{i}}$ we assume a flow rule, for the symmetric part of the inelastic strain rate tensor $\mathbf{D}_{\mathbf{0}}^{\mathbf{i}}$ in the intermediate configuration, in accordance with the flow rule proposed by Amin et al. [22]:

$$\mathbf{D}_{\mathbf{0}}^{\mathbf{i}} = \frac{1}{\eta}\left(\mathbf{P}_{\mathbf{0}} - \mathbf{P}_{\mathbf{0}}^{\mathbf{A}}\right) \tag{10}$$

where $\mathbf{P}_{\mathbf{0}}, \mathbf{P}_{\mathbf{0}}^{\mathbf{A}}$ are the so-called Mandel stress tensors [28], defined as:

$$\mathbf{P}_{\mathbf{0}} = \mathbf{F}^{\mathbf{e}^{\mathrm{T}}}\boldsymbol{\sigma}'\mathbf{F}^{\mathbf{e}^{\mathrm{T}^{-1}}} \tag{11}$$

$$\mathbf{P}_{\mathbf{0}}^{\mathbf{A}} = \mathbf{F}^{\mathbf{e}^{\mathrm{T}}}\boldsymbol{\alpha}\mathbf{F}^{\mathbf{e}^{\mathrm{T}^{-1}}} \tag{12}$$

where $\boldsymbol{\sigma}'$ is the deviatoric Cauchy stress tensor. Equation (10) represents a generalization of a Newtonian fluid, with η being a nonlinear viscosity. This expression is written in analogy to the flow rule for the inelastic strain rate tensor in the work by Huber et al. [23]. A similar flow rule is also applied in a work by Anand et al. [31]. Parameter η in this work [31] was taken to be stress and pressure dependent, so that effects related with the macroscopic response of an amorphous glassy polymer, such as yielding and strain softening could be modeled. In the work by Amin et al. [22], a power law function for the viscosity is incorporated in the constitutive equation for the inelastic strain rate tensor. The stress and strain dependence of the viscosity is modeled in terms of two parameters. In our work, a simpler form for the nonlinear viscosity η will be adapted, through an Eyring type equation:

$$\eta = \eta_0 \exp[-\delta\|\boldsymbol{\sigma}' - \boldsymbol{\alpha}\|] \tag{13}$$

where η_0 is a constant. The strain dependence of η could be considered through parameter δ, but in our calculations δ was treated as a constant.

Moreover, it is assumed that the time evolution of back stress tensor $\boldsymbol{\alpha}$ is expressed by Ziegler's law [32]:

$$\overset{\mathbf{0}}{\boldsymbol{\alpha}} = h(\boldsymbol{\alpha})\mathbf{D}^{\mathbf{i}}_{\mathbf{0}} \tag{14}$$

In our analysis, function h was taken to be stress-dependent as follows:

$$h = h_0 \exp(-\beta\|\boldsymbol{\alpha}\|) \tag{15}$$

where h_0,β are constants and $\|\boldsymbol{\alpha}\| = \sqrt{\boldsymbol{\alpha}\cdot\boldsymbol{\alpha}}$ denotes the magnitude of a tensor.

Regarding the constitutive description of the inelastic spin $\mathbf{W}^{\mathbf{i}}_{\mathbf{0}}$, a general form of constitutive equation of it may be of the type:

$$\mathbf{W}^{\mathbf{i}}_{\mathbf{0}} = \lambda\Omega^{\mathrm{i}}(\boldsymbol{\sigma}, \boldsymbol{\alpha}) \tag{16}$$

where λ is a proportional factor, and Ω^{i} is an antisymmetric tensor, and isotropic function of stress tensor $\boldsymbol{\sigma}$ and back stress tensor $\boldsymbol{\alpha}$. Following Dafalias [26] the following simplified form for Eq. 16 is proposed:

$$\mathbf{W}^{\mathbf{i}}_{\mathbf{0}} = \frac{\rho}{2}\left(\boldsymbol{\alpha}\mathbf{D}^{\mathbf{i}}_{\mathbf{0}} - \mathbf{D}^{\mathbf{i}}_{\mathbf{0}}\boldsymbol{\alpha}\right) \tag{17}$$

where ρ is a material function.

The constitutive law for $\mathbf{D}^{\mathbf{e}}$ is related with hypoelasticity theory, and taking into account that the material rate of stress tensors, involved in hypoelasticity, is not objective, the corotational rates of all types of stress tensors need to be specified. There have been extensive studies on the undesirable oscillatory responses of the stresses predicted by the improper use of the rate-type constitutive equations. Among the various proposed objective rates, the one based on the concept of plastic spin by Dafalias [25, 26] will be applied. Then the following set of constitutive equations will be used to describe the viscoelastic response at large deformations:

$$\mathbf{D}^{\mathbf{e}} = \mathrm{L}^{-1} : \overset{\mathbf{0}}{\boldsymbol{\sigma}} \tag{18}$$

$$\overset{\mathbf{0}}{\boldsymbol{\sigma}} = \dot{\boldsymbol{\sigma}} - \boldsymbol{\omega}\boldsymbol{\sigma} + \boldsymbol{\sigma}\boldsymbol{\omega} \tag{19}$$

$$\overset{\mathbf{0}}{\boldsymbol{\alpha}} = \dot{\boldsymbol{\alpha}} - \boldsymbol{\omega}\boldsymbol{\alpha} + \boldsymbol{\alpha}\boldsymbol{\omega} \tag{20}$$

where L is a fourth order tensor, $\overset{\mathbf{0}}{\boldsymbol{\sigma}}, \overset{\mathbf{0}}{\boldsymbol{\alpha}}$ are the corotational rates of Cauchy and back stress tensor, respectively, and $\boldsymbol{\omega}$ is the substructural spin.

Application to simple shear monotonic and cyclic loading

The present model will be justified on simple shear monotonic and cyclic loading performed on HDR in a recent work by Amin et al. [22]. If the problem is considered to be a two-dimensional one, the corresponding symmetric parts of the velocity gradient tensor are as follows:

$$\mathbf{D} = \begin{pmatrix} 0 & \mathrm{D}_{12} \\ \mathrm{D}_{12} & 0 \end{pmatrix} \quad \mathbf{D}^{\mathbf{e}} = \begin{pmatrix} \mathrm{D}^{\mathrm{e}}_{11} & \mathrm{D}^{\mathrm{e}}_{12} \\ \mathrm{D}^{\mathrm{ei}}_{12} & \mathrm{D}^{\mathrm{e}}_{22} \end{pmatrix} \quad \mathbf{D}^{\mathbf{i}} = \begin{pmatrix} \mathrm{D}^{\mathrm{i}}_{11} & \mathrm{D}^{\mathrm{i}}_{12} \\ \mathrm{D}^{\mathrm{i}}_{12} & \mathrm{D}^{\mathrm{i}}_{22} \end{pmatrix} \tag{21}$$

where

$$\mathrm{D}_{12} = \frac{\dot{\gamma}}{2} \tag{22}$$

and $\dot{\gamma}$ is the rate of shear deformation. The inelastic spin tensor will accordingly given by:

$$\mathbf{W}^{\mathrm{i}}_{0} = \begin{pmatrix} 0 & \mathrm{W}^{\mathrm{i}}_{0_{12}} \\ -\mathrm{W}^{\mathrm{i}}_{0_{12}} & 0 \end{pmatrix} \tag{23}$$

where its constitutive law is given by Eq. 17.

Combining Eqs. 9, 17, and 20 for monotonic shear deformation, we obtain:

$$\dot{\alpha}_{11} = h\ \mathrm{D}^{\mathrm{i}}_{0_{11}} + \alpha_{12}\dot{\gamma} + 2\rho\alpha^2_{12}\mathrm{D}^{\mathrm{i}}_{0_{11}} - 2\rho\alpha_{11}\alpha_{12}\mathrm{D}^{\mathrm{i}}_{0_{12}} \tag{24a}$$

$$\dot{\alpha}_{12} = h\ \mathrm{D}^{\mathrm{i}}_{0_{12}} - \alpha_{11}\dot{\gamma} + 2\rho\alpha^2_{11}\mathrm{D}^{\mathrm{i}}_{0_{12}} - 2\rho\alpha_{11}\alpha_{12}\mathrm{D}^{\mathrm{i}}_{0_{11}} \tag{24b}$$

Moreover, applying Eqs. 1, 3, 4, 7, and making the further assumption that small elastic deformations exist, compared to the anelastic ones, therefore $\mathbf{D} = \mathbf{D}^{\mathbf{i}}$ and $\mathbf{F}^{\mathbf{e}}$ is the identity tensor, we have:

$$\mathrm{D}^{\mathrm{i}}_{0_{11}} = 0 \quad \text{and} \quad \mathrm{D}^{\mathrm{i}}_{0_{12}} = \frac{\dot{\gamma}}{2} \tag{25}$$

and

$$\sigma'_{11} = \alpha_{11} \quad \text{and} \quad \sigma'_{12} = \alpha_{12} + \eta\frac{\dot{\gamma}}{2} \tag{26}$$

where σ'_{ij} are the corresponding components of the deviatoric Cauchy stress tensor.

Combining Eqs. 5, 7, 11, 18 and 19 for monotonic shear deformation, we obtain:

$$\dot{\sigma}_{11} = \mu\ \mathrm{D}^{\mathrm{e}}_{11} + \sigma_{12}\dot{\gamma} + 2\rho\sigma_{12}\alpha_{12}\mathrm{D}^{\mathrm{i}}_{11} - 2\rho\sigma_{12}\alpha_{11}\mathrm{D}^{\mathrm{i}}_{12} \tag{27}$$

$$\dot{\sigma}_{12} = \mu\ \mathrm{D}^{\mathrm{e}}_{12} - \sigma_{11}\dot{\gamma} + \rho\sigma_{11}\alpha_{11}\mathrm{D}^{\mathrm{i}}_{12} - \rho\sigma_{11}\alpha_{12}\mathrm{D}^{\mathrm{i}}_{11} \tag{28}$$

where μ is the shear modulus of the material.

The set of Eqs. 21–24 can be solved numerically to obtain tensors $\boldsymbol{\sigma}$ and $\boldsymbol{\alpha}$.

The total stress tensor will accordingly be given by:

$$\boldsymbol{\sigma}_{\mathbf{tot}} = \boldsymbol{\sigma} + \boldsymbol{\sigma}_{\mathbf{entr}} \tag{29}$$

where $\boldsymbol{\sigma}_{\mathbf{entr}}$ is the stress tensor related with the material's entropic hardening. It is taken to be coaxial with the inelastic stretch tensor. The model of Wang and Guth [33] of rubber elasticity was applied and the principal components of $\boldsymbol{\sigma}_{\mathbf{entr}}$ take the form:

$$(\bar{\boldsymbol{\sigma}}_{\mathbf{entrt}})_{\mathrm{i}} = \frac{C_{\mathrm{R}}}{3}\sqrt{q}\left[\lambda_{\mathrm{i}}L^{-1}\left(\frac{\lambda_{\mathrm{i}}}{\sqrt{q}}\right) - \frac{1}{3}\sum_{j}\lambda_j L^{-1}\left(\frac{\lambda_j}{\sqrt{q}}\right)\right] \tag{30}$$

where C_{R} is a rubbery modulus, $\sqrt{q}$ is the tensile locking network stretch (or natural draw ratio), L^{-1} is the inverse Langevin function, and λ_{i} are the principal inelastic stretch components, obtained by the total deformation gradient $\mathbf{F}$ and given by:

$$\begin{aligned}\lambda_1 &= \frac{1}{2}\left(\gamma + \sqrt{4+\gamma^2}\right)\\ \lambda_2 &= -\frac{1}{2}\left(\gamma - \sqrt{4+\gamma^2}\right)\\ \lambda_3 &= 1\end{aligned} \tag{31}$$

Then, tensor $\bar{\boldsymbol{\sigma}}_{\mathbf{entrt}}$ must be transformed with the appropriate transformation matrix $\mathbf{R}$:

$$\boldsymbol{\sigma}_{\mathbf{entr}} = \mathbf{R}\bar{\boldsymbol{\sigma}}_{\mathbf{entr}}\mathbf{R}^{\mathrm{T}} \tag{32}$$

with $\mathbf{R} = \begin{pmatrix} \cos\theta & \sin\theta \\ -\sin\theta & \cos\theta \end{pmatrix}$ and $\tan\theta = \lambda_1$

The validity of our approach was tested by simulating the experimental results of the monotonic response of HDR, performed by Amin et al. [22]. In Fig. 1, the experimental results, for HDR, in simple shear loading, at two different strain rates, namely 0.5 and 0.05 s^{-1} are plotted, in comparison with the simulated data of our analysis. A satisfactory agreement between theory and experiment was obtained, and the rate dependence could also be modeled in an accurate way.

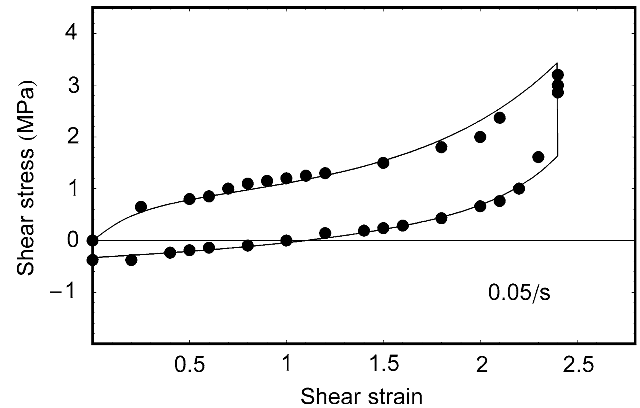

Fig. 2 Cyclic shear stress–shear strain curves at a rate of 0.05 s^{-1}. *Points*: experimental data after Amin et al. [22]. *Lines*: simulated results

Numerical calculations were made using the software Mathematica [34] for step integration of the corresponding equations.

Parameter values were estimated with a trial and error procedure, and were found to be as follows:

$h_0 = 12\ \mathrm{MPa\,s^{-1}}, \beta = 6\ \mathrm{MPa^{-1}}, \eta_0 = 600\ \mathrm{MPa\,s},$
$C_{\mathrm{R}} = 0.16\ \mathrm{MPa}, q = 3, \delta = 0.7\ \mathrm{MPa^{-1}}, \mu = 5\ \mathrm{MPa}, \rho = 1.$

The unloading response of HDR is presented in Figs. 2 and 3 at two different strain rates, after Amin et al. The strong rate dependence which is observed during loading is followed by a weak or negligible rate dependence at the unloading stage. To simulate the details of the unloading curve, parameters such as rubbery modulus C_{R} and the back-stress dependence of parameter h, namely parameter β should be modified, due to the material's structural

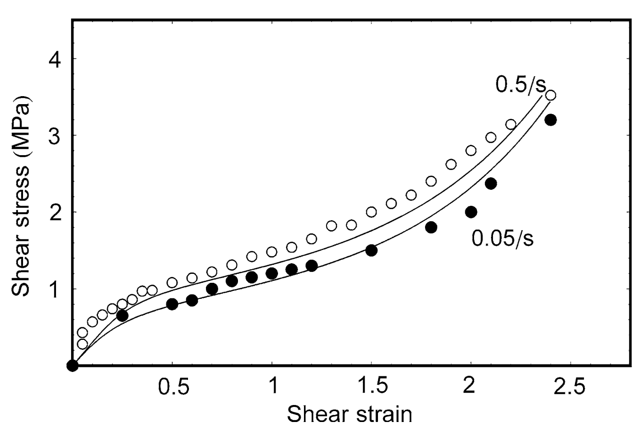

Fig. 1 Shear stress–shear strain curves at two strain rates. *Points*: experimental data after Amin et al. [22]. *Lines*: simulated results

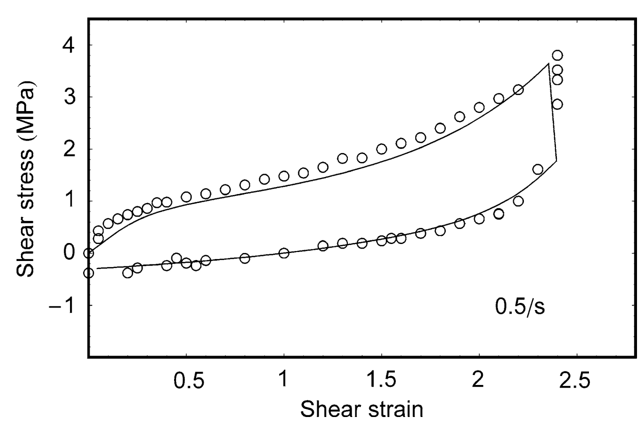

Fig. 3 Cyclic shear stress–shear strain curves at a rate of 0.5 s^{-1}. *Points*: experimental data after Amin et al. [22]. *Lines*: simulated results

changes during the first loading at large deformations. This is a reasonable assumption, given that after the first loading, a chain detachment on fillers takes place, i.e., the Mullins effect. The parameter values at the unloading procedure are as follows:

$$h_0 = 12\ \text{MPa s}^{-1}, \beta = 3\ \text{MPa}^{-1}, \eta_0 = 600\ \text{MPa s},$$
$$C_R = 0.06\ \text{MPa}, q = 3, \delta = 0.7\text{MPa}^{-1}, \mu = 5\ \text{MPa}, \rho = 1$$

In this way, it is observed from Figs. 2 and 3 that the simulated results in unloading appear the same trend as the experimental ones.

Application to stress relaxation

The validity of our model was further examined on stress relaxation tests in simple shear. For this type of deformation, the kinematics are expressed by the following equations:

$$\mathbf{D} = \mathbf{0}, \quad \mathbf{W} = \mathbf{0} \tag{33}$$

$$\mathbf{D^i} = -\mathbf{D^e} \tag{34}$$

$$\mathbf{D^i_0} = \frac{1}{\eta}(\mathbf{P_0} - \mathbf{P^A_0}) \tag{35}$$

Following Eqs. 9 and 33, we obtain for the substructural spin $\boldsymbol{\omega}$:

$$\boldsymbol{\omega} = -\mathbf{W^i_0} = -\frac{\rho}{2}\left(\boldsymbol{\alpha}\mathbf{D^i_0} - \mathbf{D^i_0}\boldsymbol{\alpha}\right). \tag{36}$$

We also have for the symmetric part $\mathbf{D^e}$ of the elastic velocity gradient that it is equal to:

$$\mathbf{D^e} = [\dot{\mathbf{F}}^{\mathbf{e}}\mathbf{F}^{\mathbf{e}^{-1}}]_s \tag{37}$$

Combining Eqs. 7, 34, 35 and 36 differential equations for the components of tensor $\dot{\mathbf{F}}^{\mathbf{e}}$ were obtained, as functions of the components of tensor $\mathbf{F^e}$. The initial values of $\mathbf{F^e}$ were taken to be compatible with the instantaneously imposed shear strain.

Moreover, from Eqs. 19 and 20 the following set of differential equations for the deviatoric back stress tensor is extracted:

$$\dot{\alpha}_{11} = h\ d^i_{0_{11}} + 2\rho\alpha^2_{12}\ d^i_{0_{11}} - 2\rho\alpha_{11}\alpha_{12}\ d^i_{0_{12}} \tag{38}$$

$$\dot{\alpha}_{12} = h\ d^i_{0_{12}} - 2\rho\alpha_{11}\alpha_{12}\ d^i_{0_{11}} + 2\rho\alpha^2_{11}\ d^i_{0_{12}} \tag{39}$$

where $d^i_{0_{ij}}$ are the components of tensor $\mathbf{D^i_0}$.

The time evolution of Cauchy stress tensor $\boldsymbol{\sigma}$ is extracted by combining Eqs. 18, 19 and 34, and thus complicated functions of the components of $\mathbf{D^e}$ and $\mathbf{F^e}$ are obtained.

The numerical calculations were performed considering Eqs. 24–28 and 35–39. The integration has been made by using small time steps until a high convergence is obtained.

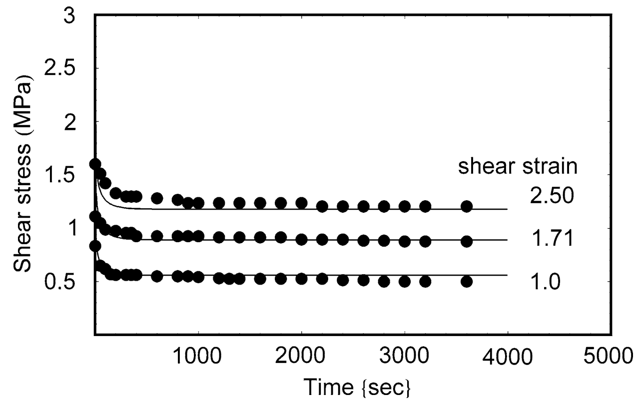

Fig. 4 Shear stress-relaxation curves at three shear strains. *Points*: experimental data after Amin et al. [22]. *Lines*: simulated results

All calculations were included in a computer program in the frame of the software Mathematica [34]. The simulated results for shear stress relaxation are presented in comparison with the experimental ones in Fig. 4. For the three shear strains examined, namely 1.0, 1.71 and 2.5, a very satisfactory agreement between theory and experiment was achieved, at both, the short-time response and the equilibrium state at long times as well. The calculations for stress-relaxation were performed with the same set of parameter values applied for monotonic loading.

Conclusions

The nonlinear viscoelastic response of materials at large deformations is formulated through a constitutive approach, valid in plasticity, which includes the fundamental principles of thermodynamic consistency, objectivity of kinematic quantities as well as the determinance of the intermediate (inelastic) configuration, being assured by the substructural constitutive spin $\boldsymbol{\omega}$. Nonlinear effects of rubbers, expressed by rate dependence, cyclic loading and stress relaxation tests were captured through the introduction of the constitutive spin, the back stress tensor and a nonlinear viscosity which is stress dependent. This approach is a contribution to the real complex nonlinear viscoelastic response of rubbers, which appear a variety of relaxation mechanisms, and consequently a variation of viscosity coefficients. In those time scales, where a weak dependence of the viscosity on rate, stress and strain is expected, the proposed description predicts the material's response with a high accuracy. Whenever, the viscosity dependence on rate, stress and strain is strong, deviations between experimental data and simulated results are exhibited. To face this shortcoming, a simple stress dependent nonlinear function of the viscosity has been adapted, and proved to formulate the real material response in a satisfactory way. On the other hand, to simulate the cyclic loading, parameters related with the material structure such as the rubbery modulus C_R and parameter β, were accordingly modified, denoting this way the structure degradation occurring in the first cycle of loading.

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