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A physically based method to represent the thermo-mechanical behaviour of elastomers

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Summary. A geometric nonlinear and thermodynamical consistent constitutive theory is proposed, which allows the representation of the thermomechanical behaviour of carbon black filled rubber. In a recent paper [1] it was shown that the mechanical behaviour of this material is mainly influenced by nonlinear elasticity coupled with some inelastic effects, in particular the Mullins-effect, nonlinear rate dependence and a weak equilibrium hysteresis. In the present paper, the Mullins-effect is not taken into consideration. At first we discuss a uniaxial approach, based on a simple spring dashpot system of viscoplasticity. The essential feature of this model is a decomposition of the total stress into a rate independent equilibrium stress and a nonlinear rate dependent overstress. The equilibrium stress is decomposed into a sum of two terms as well: The first term, the elastic part of the equilibrium stress, is a nonlinear function of the total strain, and the second term, the so-called hysteretic part, depends in a rate independent manner on the strain history. Both the overstress and the hysteretic part of the equilibrium stress are determined by nonlinear elasticity relations which depend on internal variables. These internal variables are inelastic strains, and the corresponding evolution equations are developed in consideration of the second law of thermodynamics. Accordingly, we demonstrate that the principle of non-negative dissipation is satisfied for arbitrary deformation processes. In a further step, we transfer the structure of this model to the three-dimensional and geometric nonlinear case. In a certain sense similar to finite deformation elasto-plasticity, we introduce two multiplicative decompositions of the deformation gradient into elastic and inelastic parts. The first decomposition is defined with respect to the overstress and the second one with respect to the hysteretic part of the equilibrium stress. Consequently, two intermediate configurations are induced, which lead two different decompositions of the Green's strain tensor into elastic and inelastic parts. The latter are the internal variables of the model. For physical reasons, we define the corresponding stress tensors and derivatives in the sense of the concept of dual variables [7], [39]. The constitutive equations for the overstress and for the hysteretic part of the equilibrium stress are specified by nonlinear elasticity relations, formulated with respect to the different intermediate configurations. In order to facilitate a separate description of inelastic bulk and distortional effects, we introduce kinematic decompositions of the deformation gradient into volumetric and distortional parts. Numerical simulations demonstrate that the developed theory represents the mechanical behaviour of a tread compound at room temperature very well. Thermomechanical heating effects, which are caused by inelastic deformations are also described by the theory. The method proposed in this paper can be utilised to generalise uniaxial rheological models to three-dimensional finite strain theories, which are admissible in the sense of the second law of thermodynamics.

1 Introduction

A quite interesting problem in rubber engineering is the optimisation of energy dissipation in rolling tyres. To this end constitutive theories are required, which allow the mathematical representation of the inelastic behaviour of filler loaded rubbers in combination with thermomechanical heating effects. From experience it is known, that each material which is loaded from the virgin state by inelastic deformation heats up.

A certain part of the stress power is stored in the interior of the material, and the rest is dissipated into heat. If the material properties depend on temperature, the temperature rise influences the stress response behaviour and the dissipation properties. Accordingly, it stands to reason that physical reasonable constitutive theories should be able to represent both the temperature dependent mechanical behaviour and the dissipation properties. Examined from a point of view of mathematics, this concept leads in general to thermomechanically coupled boundary value problems. Computational methods to solve such problems are developed for example by Miehe [23], [26].

In the recent past, there are proposed numerous models for the mathematical representation of the elastic and the inelastic behaviour of rubberlike materials. The elastic phenomena of natural rubber are already known and have been investigated by many researchers in the past. Some basic results are well reported by James et al. [32] where this kind of material behavior is sufficiently represented by classical models of finite deformation hyperelasticity, i.e. by the classical Mooney-Rivlin theory, or by generalised Neo-Hookean models. In order to represent the Mullins-effect and the high-cycle fatigue behaviour of rubber in combination with nonlinear elasticity, Miehe proposes an Ogden-type model in combination with two damage variables [24]. Physically, these variables are attributed to the different damage mechanisms. In contrast to this approach, Govindjee and Simo propose a micro-mechanically based damage concept [28]. Up to now many methods mainly based on elasticity (James and Green [33]) or viscoelasticity (Christensen [37], Lubliner [4], Simo [27], Hausler and Sayir [36]), have been developed to describe the mechanical behaviour of filler loaded rubbers. For example, James and Green [33] neglect the rate dependent part of the stress after some recovery time and utilise a strain energy function to describe the equilibrium behavior. In contrast to these approaches, Browning et al. [35] apply a uniaxial model of viscoplasticity to represent the mechanical behaviour of a filled polymer. A quite interesting thermomechanical model of finite deformation viscoelasticity has been developed by Holzapfel [29] and by Holzapfel and Simo [30]. The theory is compatible with the second law of thermodynamics in form of the Clausius Duhem inequality, but nonlinear rate dependence and equilibrium hysteresis effects are not incorporated.

The main objective of the present work is to develop a thermodynamical consistent internal variable theory, which allows the representation of the inelastic behaviour of filler loaded rubbers. In particular, the model must incorporate nonlinear elasticity and nonlinear rate dependence in combination with equilibrium hysteresis effects. In addition, the second law of thermodynamics has to be satisfied for arbitrary thermomechanical loadings, i.e. the rate of the specific entropy production γ must be non-negative for any deformation- and temperature-process:

$$\gamma \geq 0 \tag{1}$$

A further topic of this paper is the representation of thermomechanical heating effects, which are caused by inelastic deformations. Typically, these phenomena appear during cyclic deformations with high loading frequencies, for example in rolling tyres [21]. Experimental results discussed in [1] show the mechanical behaviour of a filler loaded tread compound at room temperature to be mainly influenced by nonlinear elasticity coupled with the Mullins-effect, nonlinear rate dependence and a weak equilibrium hysteresis. In particular, the Mullins-effect can be attributed to the breakage of weak bonds between the rubber matrix and filler particles during deformation [2], [3]. Since this effect occurs only during the first periods of the deformation process, it has no influence on the material's response during stationary cyclic loadings. Phenomenologically, this phenomenon can be represented by damage variables (see Lion [1], Miehe [24], Simo [27]). Rate dependence is observed during monotonic and cyclic tests in tension and compression under different values of the strain rate: In general, the stress increases monotonically and strongly nonlinear with increasing strain rates. The third inelastic phenomenon, the so-called equili-brium hysteresis, can be observed during cyclic loadings in combination with a series of relaxation periods with sufficient long duration [1]. In terms of the micro structure, this effect can be attributed to irreversible slip processes between adjacent filler particles (Kilian et al. [2]). In this context, Turner [13] has developed a uniaxial rate independent friction model, which describes the equilibrium hysteresis behaviour in combination with the corresponding energy loss mechanisms in filler reinforced rubber very well. Summarising, rate dependence in combination with equilibrium hysteresis leads, to energy dissipation in combination with thermomechanical heating effects. In order to develop the constitutive theory, some basic ideas were taken from Lubliner [4], Haupt [5] and Haupt and Tsakmakis [7], [39].

2 Uniaxial approach

The objective of the following Section is to design a reasonable uniaxial constitutive model, which provides a qualitative description of the mechanical behaviour of rubber, i.e. nonlinear elasticity coupled with equilibrium hysteresis and nonlinear rate dependence. For physical reasons, the model is based on a rheological spring-dashpot system. The basic advantage of this method is the direct relation of the experimentally observed phenomena. In addition, the corresponding Helmholtz free energy can easily be derived, and thermomechanical consistency can be demonstrated (see Krawietz [19]). The basic ideas of the model are additive stress- and corresponding strain decompositions. In the third Section, we generalise the structure of this model to the three-dimensional case at finite strain. To this end, we introduce two multiplicative decompositions of the deformation gradient into elastic and inelastic parts. Then, we apply the concept of dual variables [7] and specify a set of physically reasonable strain- and stress measures. It is shown that the three-dimensional theory is thermodynamically consistent as well. In the theory of finite deformation rubber viscoelasticity, the idea to decompose the deformation gradient multiplicatively was introduced by Lubliner [4]. As proposed by Malmberg [12], the presented theory is developed right from the beginning in consideration of thermodynamical aspects.

Now we discuss the uniaxial rheological model. The basic ideas are additive stress- and strain decompositions, which are illustrated in Fig. 1. The spring in the centre of the model is introduced to represent the nonlinear elastic behaviour. The Maxwell element at the bottom is attributed to the rate dependent part of the stress and the friction element at the top describes the equilibrium hysteresis effects. All springs are assumed to be nonlinear functions of the corresponding strain variables ε_{ep}, ε, and ε_{ev}.

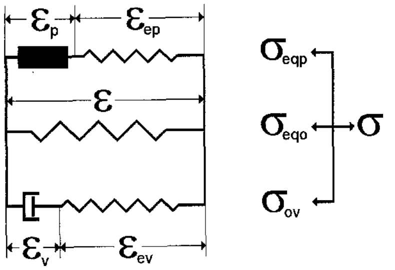

Fig. 1. One-dimensional rheological model of viscoplasticity

The dashpot at the bottom stands for a nonlinear viscosity, and the black box at the top symbolises a rate independent friction element without any yield condition. From a point of view of physics, the introduction of a distinct yield stress like in elasto-plasticity seems to be not reasonable in case of rubberlike materials. As shown in Fig. 1, the total stress σ decomposes additively into a rate independent equilibrium stress and a rate dependent overstress σ_{ov}. The equilibrium stress itself is a sum of two terms: The first term σ_{eqo} is a nonlinear function of the total strain ε, and the second term σ_{eqp} is a rate independent functional of the strain history,

$$\sigma = \sigma_{eqo}(\varepsilon) + \sigma_{eqp}(\varepsilon_{ep}) + \sigma_{ov}(\varepsilon_{ev}). \tag{2}$$

In addition to the stress decomposition, the strain decompositions are now introduced. Figure 1 motivates two different decompositions into elastic and inelastic parts. They are defined by

$$\varepsilon = \varepsilon_{ev} + \varepsilon_v \quad \text{and} \quad \varepsilon = \varepsilon_{ep} + \varepsilon_p. \tag{3}$$

The first decomposition is related to the overstress, and ε_v is the inelastic strain of the nonlinear dashpot, whereas the second decomposition corresponds to the hysteretic part of the equilibrium stress, and ε_p is the inelastic strain of the friction element. For physical reasons we introduce three strain energy functions, which are related to the springs. In particular, the function w_0 depends on the total strain ε, w_v is a function of the elastic strain ε_{ev} of the Maxwell element, and the strain energy function w_p depends on the elastic strain ε_{ep} of the friction element.

$$w_0 = w_0(\varepsilon), \qquad w_p = w_p(\varepsilon_{ep}), \qquad w_v = w_v(\varepsilon_{ev}). \tag{4}$$

Now, we specify the constitutive relations for the stress quantities and the flow rules for the inelastic strains. For this purpose, we follow Malmberg [12] and take the second law of thermodynamics into consideration. In the present paper, the term *thermodynamical consistency* means compatibility with the Clausius Duhem inequality. The constitutive relations are formulated so that the rate of the specific entropy production γ is non-negative for arbitrary temperature- and deformation processes (see [8]). In order to fix the main ideas, we start with the investigation of isothermal deformation processes. For these processes, the uniaxial form of the specific entropy production reads

$$\varrho\gamma = -\varrho\dot{\psi} + \sigma\dot{\varepsilon}, \tag{5}$$

where ϱ is the mass density, ψ the Helmholtz free energy per unit mass and $\sigma\dot{\varepsilon}$ the stress power per unit volume. The Helmholtz energy of the rheological model in Fig. 1 is given by the total energy, stored in the springs:

$$\psi = w_0(\varepsilon) + w_p(\varepsilon_{ep}) + w_v(\varepsilon_{ev}). \tag{6}$$

A comparison between Eqs. (2) and (6) shows that the free energy decomposes in the same manner as the stress. If we insert the constitutive relations, given by Eqs. (2), (3), and (6), into Eq. (5) we obtain the following expression for the specific entropy production:

$$\varrho\gamma = \left(\sigma_{eqo} - \varrho\,\frac{\partial w_o}{\partial \varepsilon}\right)\dot{\varepsilon} + \left(\sigma_{eqp} - \varrho\,\frac{\partial w_p}{\partial \varepsilon_{ep}}\right)\dot{\varepsilon} + \left(\sigma_{ov} - \varrho\,\frac{\partial w_v}{\partial \varepsilon_{ev}}\right)\dot{\varepsilon} + \varrho\,\frac{\partial w_p}{\partial \varepsilon_{ep}}\,\dot{\varepsilon}_p + \varrho\,\frac{\partial w_v}{\partial \varepsilon_{ev}}\,\dot{\varepsilon}_v. \tag{7}$$

In Eq. (7) all coefficients of the total strain rate $\dot{\varepsilon}$ are zero, because the springs are assumed to be hyperelastic:

$$\sigma_{eqo} = \varrho \frac{\partial w_0}{\partial \varepsilon}, \quad \sigma_{eqp} = \varrho \frac{\partial w_p}{\partial \varepsilon_{ep}}, \quad \sigma_{ov} = \varrho \frac{\partial w_v}{\partial \varepsilon_{ev}}. \tag{8}$$

The remaining terms in Eq. (7) show that the choice of the evolution equations for the inelastic strains ε_p and ε_v cannot be arbitrary if the model is required to be compatible with the dissipation inequality. Consequently, we define the flow rules by the formulae

$$\dot{\varepsilon}_p = \dot{z} \frac{\varrho}{\eta_p} \frac{\partial w_p}{\partial \varepsilon_{ep}} \quad \text{and} \quad \dot{\varepsilon}_v = \frac{\varrho}{\eta_v} \frac{\partial w_v}{\partial \varepsilon_{ev}} \quad \text{with} \quad \eta_v > 0 \quad \text{and} \quad \eta_p > 0, \tag{9}$$

so that the driving force for the viscous strain ε_v is the overstress and for the plastic strain ε_p the hysteretic part of the equilibrium stress. Motivated by Fig. 1, the material functions η_v and η_p may depend, if necessary, on the arguments σ_{ov} or ε_v and σ_{eqp} or ε_p, respectively. Other dependencies, for example $\eta_v = \eta_v(\sigma)$ or $\eta_v = \eta_v(\varepsilon)$, are physically not reasonable because neither the total stress σ nor the total strain ε is acting directly on the corresponding element. The non-negative quantity $\dot{z}$ in Eq. (9) is the time rate of the kinematic arclength, $\dot{z} = |\dot{\varepsilon}|$, or an arclength of a generalised type (see for example Valanis [9] or Haupt and Lion [6]). Then, the plastic strain ε_p depends in a rate independent manner on the deformation history. A simple conclusion resulting from the flow rules is that the dissipated stress power of the friction element and the dashpot is non-negative for arbitrary deformation processes:

$$\sigma_{eqp}\dot{\varepsilon}_p = \dot{z} \frac{1}{\eta_p} \left(\varrho \frac{\partial w_p}{\partial \varepsilon_{ep}} \right)^2 \geq 0, \quad \sigma_{ov}\dot{\varepsilon}_v = \frac{1}{\eta_v} \left(\varrho \frac{\partial w_v}{\partial \varepsilon_{ev}} \right)^2 \geq 0. \tag{10}$$

At this point, the construction of the uniaxial and thermodynamically admissible model of viscoplasticity is completed. The model incorporates nonlinear elasticity, rate-dependence and equilibrium hysteresis. The basic ideas are additive stress decompositions in combination with corresponding strain decompositions. The flow rules for the inelastic strains are specified in consideration of the Clausius Duhem inequality.

3 Finite strain theory

As shown in the previous Section, the rheological model incorporates all experimentally observed phenomena and is thermodynamically consistent. Motivated by these properties, we generalise the basic ideas as far as possible to obtain a three-dimensional theory of viscoplasticity. The ingredients of the uniaxial model are stress- and strain decompositions, a free energy function depending on the elastic parts of the strain and flow rules for the inelastic strains. A well known problem connected with the development of finite deformation models for inelastic material behaviour is the definition of reasonable strain decompositions in combination with associated stress tensors. This problem has been discussed at full length for example by Haupt and Tsakmakis [7], [39], where the concept of dual variables and derivatives was developed. In the framework of finite deformations, it is a common practice to start out from a multiplicative decomposition of the deformation gradient into elastic and inelastic parts, like in elasto-plasticity (see for example Besdo [10], Miehe [26], or Haupt and Tsakmakis [7]). Lubliner [4] has applied this concept to finite strain viscoelasticity and Boyce et al. [22] have utilised this idea to represent the inelastic behaviour of polymers. Stickforth discusses some physical implications of such decompositions [38].

3.1 Fundamentals

In this Subsection, we provide a set of physically reasonable stress- and strain measures, which may be utilised to formulate three-dimensional constitutive equations. First of all, we discuss the strain decompositions. By the rheological model in Fig. 1, two additive decompositions are defined (Eqs. (3)). The first one is related to the rate dependent overstress and the second one to the rate independent hysteretic part of the equilibrium stress. In order to transfer this idea to the three-dimensional case, we introduce two different decompositions of the deformation gradient $\boldsymbol{F}$ into elastic and inelastic parts. They are defined by the relations

$$\boldsymbol{F} = \boldsymbol{F}_{ev}\boldsymbol{F}_v \quad \text{and} \quad \boldsymbol{F} = \boldsymbol{F}_{ep}\boldsymbol{F}_p \tag{11}$$

and are illustrated in Fig. 2. $\mathscr{R}$ is the reference configuration and $\mathscr{C}$ is the current configuration. The intermediate configurations, which are generated by these decompositions are denoted by $\mathscr{F}_v$ and $\mathscr{F}_p$.

The first decomposition of Eq. (11) is related to the overstress, and the inelastic part of the deformation gradient is denoted by $\boldsymbol{F}_v$, whereas the second decomposition is attributed to the hysteretic part of the equilibrium stress, and the inelastic part of $\boldsymbol{F}$ is denoted by $\boldsymbol{F}_p$. On this basis, we can formulate two physically reasonable strain measures which allow an additive split into elastic and inelastic parts. If we insert both decompositions into the common definition of the Green's strain tensor $\boldsymbol{E} = 1/2\ (\boldsymbol{F}^T\boldsymbol{F} - \boldsymbol{1})$, we obtain the following expressions:

$$\boldsymbol{E} = \frac{1}{2}\,(\boldsymbol{F}_v{}^T(\boldsymbol{F}_{ev}^T\boldsymbol{F}_{ev})\,\boldsymbol{F}_v - \boldsymbol{1}), \qquad \boldsymbol{E} = \frac{1}{2}\,(\boldsymbol{F}_p{}^T(\boldsymbol{F}_{ep}^T\boldsymbol{F}_{ep})\,\boldsymbol{F}_p - \boldsymbol{1}). \tag{12}$$

By pushing forward Eqs. (12) to the configurations $\mathscr{F}_v$ and $\mathscr{F}_p$, we obtain the strain tensors ε_{ov} and ε_{eqp}:

$$\varepsilon_{ov} := \boldsymbol{F}_v{}^{T-1}\boldsymbol{E}\boldsymbol{F}_v{}^{-1} = \frac{1}{2}\,(\boldsymbol{F}_{ev}^T\boldsymbol{F}_{ev} - \boldsymbol{1}) + \frac{1}{2}\,(\boldsymbol{1} - \boldsymbol{F}_v{}^{T-1}\boldsymbol{F}_v{}^{-1}) =: \varepsilon_{ev} + \varepsilon_v, \tag{13.1}$$

$$\varepsilon_{eqp} := \boldsymbol{F}_p{}^{T-1}\boldsymbol{E}\boldsymbol{F}_p{}^{-1} = \frac{1}{2}\,(\boldsymbol{F}_{ep}^T\boldsymbol{F}_{ep} - \boldsymbol{1}) + \frac{1}{2}\,(\boldsymbol{1} - \boldsymbol{F}_p{}^{T-1}\boldsymbol{F}_p{}^{-1}) =: \varepsilon_{ep} + \varepsilon_p. \tag{13.2}$$

Clearly, each of them decomposes additively into a purely elastic and a purely inelastic part. The elastic parts are denoted by ε_{ev} and ε_{ep} and the inelastic parts by ε_v and ε_p. Physically, these

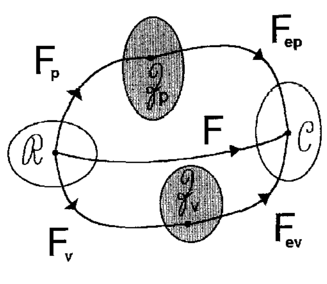

Fig. 2. Two different decompositions of the deformation gradient

decompositions are related to the Maxwell element (Eq. (13.1)) and to the rate independent friction element (Eq. (13.2)). In the next Section we utilise these strain tensors to transfer the rheological model to finite strain. The next stage is the definition of physically reasonable stress measures which are associated to these strain tensors.

By the rheological model a split of the total stress into a rate dependent overstress and rate independent equilibrium stress was introduced. The equilibrium stress was a sum of a nonlinear function of the total strain and a history dependent part. In the following, we assume the same decomposition for the second Piola Kirchhoff stress tensor $\tilde{\boldsymbol{T}}$:

$$\tilde{\boldsymbol{T}} = \tilde{\boldsymbol{T}}_{eqo} + \tilde{\boldsymbol{T}}_{eqp} + \tilde{\boldsymbol{T}}_{ov}. \tag{14}$$

Accordingly, this stress decomposition is associated to the reference configuration $\mathscr{R}$, whereas the strain decompositions are related to the configurations $\mathscr{F}_v$ and $\mathscr{F}_p$. For that reason, we transform the overstress $\tilde{\boldsymbol{T}}_{ov}$ to the intermediate configuration $\mathscr{F}_v$ and the hysteretic part $\tilde{\boldsymbol{T}}_{eqp}$ of the equilibrium stress to the configuration $\mathscr{F}_p$. There, they can be related by constitutive equations to their associated strain tensors ε_{ev} and ε_{ep}. The elastic part $\tilde{\boldsymbol{T}}_{eqo}$ of the equilibrium stress does not need to be transformed, because the associated strain tensor $\boldsymbol{E}$ operates on the reference configuration too. First of all, we carry out the transformations for which we apply the concept of dual variables [7]:

$$\tau_{ov} := \boldsymbol{F}_v \tilde{\boldsymbol{T}}_{ov} \boldsymbol{F}_v{}^T, \qquad \tau_{eqp} := \boldsymbol{F}_p \tilde{\boldsymbol{T}}_{eqp} \boldsymbol{F}_p{}^T. \tag{15}$$

The tensors τ_{ov} and τ_{eqp} are the overstress and the hysteretic part of the equilibrium stress, pushed forward to the corresponding intermediate configurations. In the next Subsection, we utilise these stress tensors in combination with the strain tensors of Eqs. (13) to design the constitutive model. In addition, we need the associated rates of these variables. Within the concept of dual variables, the same transformations as utilised in Eqs. (13) and (15) define the rates. These are the following Oldroyd derivatives:

$$\overset{\nabla}{\tau}_{ov} := \boldsymbol{F}_v \dot{\tilde{\boldsymbol{T}}}_{ov} \boldsymbol{F}_v{}^T = \dot{\tau}_{ov} - \boldsymbol{L}_v \tau_{ov} - \tau_{ov} \boldsymbol{L}_v{}^T, \tag{16.1}$$

$$\overset{\nabla}{\tau}_{eqp} := \boldsymbol{F}_p \dot{\tilde{\boldsymbol{T}}}_{eqp} \boldsymbol{F}_p{}^T = \dot{\tau}_{eqp} - \boldsymbol{L}_p \tau_{eqp} - \tau_{eqp} \boldsymbol{L}_p{}^T, \tag{16.2}$$

$$\overset{\Delta}{\varepsilon}_{ov} := \boldsymbol{F}_v^{T-1} \dot{\boldsymbol{E}} \boldsymbol{F}_v{}^{-1} = \dot{\varepsilon}_{ov} + \boldsymbol{L}_v{}^T \varepsilon_{ov} + \varepsilon_{ov} \boldsymbol{L}_v, \tag{17.1}$$

$$\overset{\Delta}{\varepsilon}_{eqp} := \boldsymbol{F}_p^{T-1} \dot{\boldsymbol{E}} \boldsymbol{F}_p{}^{-1} = \dot{\varepsilon}_{eqp} + \boldsymbol{L}_p{}^T \varepsilon_{eqp} + \varepsilon_{eqp} \boldsymbol{L}_p, \tag{17.2}$$

where the viscous and plastic deformation rates $\boldsymbol{L}_v$ and $\boldsymbol{L}_p$ are defined by

$$\boldsymbol{L}_v = \dot{\boldsymbol{F}}_v \boldsymbol{F}_v{}^{-1} \quad \text{and} \quad \boldsymbol{L}_p = \dot{\boldsymbol{F}}_p \boldsymbol{F}_p{}^{-1}. \tag{18}$$

A consequence resulting from Eqs. (13) and (17) is that the associated strain rates are also additively splitted into purely elastic and inelastic parts:

$$\overset{\Delta}{\varepsilon}_{ov} = \overset{\Delta}{\varepsilon}_{ev} + \overset{\Delta}{\varepsilon}_{v}, \qquad \overset{\Delta}{\varepsilon}_{eqp} = \overset{\Delta}{\varepsilon}_{ep} + \overset{\Delta}{\varepsilon}_{p}. \tag{19}$$

An important property of the dual variables turns out if we calculate the stress power of the overstress and the hysteretic part of the equilibrium stress with respect to both the reference and the intermediate-configuration:

$$\tilde{\boldsymbol{T}}_{ov} \cdot \dot{\boldsymbol{E}} = \tau_{ov} \cdot \overset{\Delta}{\varepsilon}_{ov} = \tau_{ov} \cdot [\overset{\Delta}{\varepsilon}_{ev} + \overset{\Delta}{\varepsilon}_{v}], \qquad \tilde{\boldsymbol{T}}_{eqp} \cdot \dot{\boldsymbol{E}} = \tau_{eqp} \cdot \overset{\Delta}{\varepsilon}_{eqp} = \tau_{eqp} \cdot [\overset{\Delta}{\varepsilon}_{ep} + \overset{\Delta}{\varepsilon}_{p}]. \tag{20}$$

Obviously, both stress power terms are invariant under the transformations discussed above. In addition, they decompose additively into elastic and inelastic parts. This property is very important in view of the thermodynamical investigation of constitutive theories. At this point, the physical and mathematical foundations are complete and the next topic is the development of the three-dimensional finite strain model.

3.2 Constitutive assumptions and the second law of thermodynamics

Now, a set of reasonable stress- and strain tensors in combination with associated rates is provided which can be utilised to generalise the rheological model to the geometrically nonlinear case. At first, we consider the Helmholtz free energy ψ, which is related to the total energy stored in the springs of the system. Physically, this property corresponds to an additive split into three terms (see Eq. (6)): The first term depends on the current value of the total strain, whereas the second and the third term depend on the strain history. In particular, the second term is the energy stored in the friction element and the third term is the energy stored in the Maxwell element. In the following, we assume that this concept is also valid in the three-dimensional non-isothermal case. Straightforward generalisation of Eq. (6) leads to the following ansatz for the free energy per unit mass:

$$\psi = w_0(\boldsymbol{E}, \theta) + w_p(\varepsilon_{ep}, \theta) + w_v(\varepsilon_{ev}, \theta) + \varphi(\theta). \tag{21}$$

In Eq. (21), the variable θ denotes the absolute temperature and $\varphi(\theta)$ is related to the specific heat capacity per unit mass. The function w_0 depends on the Green's strain $\boldsymbol{E}$ and on θ, whereas w_p and w_v are assumed to depend on temperature and on the elastic strains ε_{ep} and ε_{ev} operating on the intermediate configurations $\mathscr{F}_p$ and $\mathscr{F}_v$. Motivated by the rheological model, we decompose the second Piola Kirchhoff stress tensor $\tilde{\boldsymbol{T}}$ in the same manner as the free energy function (see Haupt [5]):

$$\tilde{\boldsymbol{T}} = \tilde{\boldsymbol{T}}_{eqo} + \tilde{\boldsymbol{T}}_{eqp} + \tilde{\boldsymbol{T}}_{ov}.$$

The next step is the specification of the constitutive relations for the three stress tensors and the specification of the flow rules for the inelastic strain tensors ε_p and ε_v. A quite important result of the uniaxial approach was that the flow rules cannot be chosen arbitrarily if the constitutive model is required to be compatible with the second law of thermodynamics. To this end, we investigate the general form of the specific entropy production

$$\varrho_R \gamma = -\varrho_R \dot{\psi} + \tilde{\boldsymbol{T}} \cdot \dot{\boldsymbol{E}} - \varrho_R s \dot{\theta} - \frac{\boldsymbol{q}_R \cdot \boldsymbol{g}_R}{\theta} \tag{22}$$

under the constitutive assumptions existing hitherto. In Eq. (22), the variable s denotes the specific entropy per unit mass, $\boldsymbol{g}_R$ is the temperature gradient with respect to the reference configuration, ϱ_R is the corresponding mass density and $\boldsymbol{q}_R$ the heat flux. Inserting the ansatz for the Helmholtz free energy in combination with the stress decomposition into Eq. (22), we obtain the following expression:

$$\begin{aligned} \varrho_R \gamma = {} & \left(\tilde{\boldsymbol{T}}_{eqo} - \varrho_R \frac{\partial w_0}{\partial \boldsymbol{E}} \right) \cdot \dot{\boldsymbol{E}} - \varrho_R \left(s + \frac{\partial w_0}{\partial \theta} + \frac{\partial w_p}{\partial \theta} + \frac{\partial w_v}{\partial \theta} + \frac{\partial \varphi}{\partial \theta} \right) \dot{\theta} - \frac{\boldsymbol{q}_R \cdot \boldsymbol{g}_R}{\theta} \\ & + \tilde{\boldsymbol{T}}_{eqp} \cdot \dot{\boldsymbol{E}} + \tilde{\boldsymbol{T}}_{ov} \cdot \dot{\boldsymbol{E}} - \varrho_R \frac{\partial w_p}{\partial \varepsilon_{ep}} \cdot \dot{\varepsilon}_{ep} - \varrho_R \frac{\partial w_v}{\partial \varepsilon_{ev}} \cdot \dot{\varepsilon}_{ev} \end{aligned} \tag{23}$$

In order to state physically reasonable and sufficient conditions for the compatibility with the dissipation principle of thermodynamics, we set the first term in brackets at zero. Accordingly, the elastic part $\tilde{\boldsymbol{T}}_{eqo}$ of the equilibrium stress is specified by the following relation of hyperelasticity:

$$\tilde{\boldsymbol{T}}_{eqo}(\boldsymbol{E}, \theta) = \varrho_R \frac{\partial w_0}{\partial E}. \tag{24}$$

In the case of natural rubber the dependence of the potential $w_0(\boldsymbol{E}, \theta)$ on temperature is homogeneous and linear (see Treloar [11], Miehe [25], or Holzapfel [29]). In order to satisfy the dissipation principle for arbitrary temperature processes, we set the coefficient of the temperature rate at zero and obtain the expression

$$s = -\left(\frac{\partial w_0}{\partial \theta} + \frac{\partial w_p}{\partial \theta} + \frac{\partial w_v}{\partial \theta} + \frac{\partial \varphi}{\partial \theta}\right) \tag{25}$$

for the specific entropy s. Now, we replace the inelastic stress power terms of Eq. (23) by Eqs. (20) and express the time rates of the elastic strains ε_{ev} and ε_{ep} in terms of their corresponding Oldroyd derivatives (Eqs. (17) and (19)):

$$\dot{\varepsilon}_{ev} = \overset{\triangle}{\varepsilon}_{ev} - \boldsymbol{L}_v{}^T \varepsilon_{ev} - \varepsilon_{ev} \boldsymbol{L}_v \quad \text{and} \quad \dot{\varepsilon}_{ep} = \overset{\triangle}{\varepsilon}_{ep} - \boldsymbol{L}_p{}^T \varepsilon_{ep} - \varepsilon_{ep} \boldsymbol{L}_p. \tag{26}$$

After inserting Eqs. (26) into the specific entropy production and rearranging terms, we obtain the result

$$\begin{aligned} \varrho_R \gamma = {} & \left(\boldsymbol{\tau}_{ov} - \varrho_R \frac{\partial w_v}{\partial \varepsilon_{ev}}\right) \cdot \overset{\triangle}{\varepsilon}_{ev} + \left(\boldsymbol{\tau}_{eqp} - \varrho_R \frac{\partial w_p}{\partial \varepsilon_{ep}}\right) \cdot \overset{\triangle}{\varepsilon}_{ep} - \frac{\boldsymbol{q}_R \cdot \boldsymbol{g}_R}{\theta} + \varrho_R \frac{\partial w_p}{\partial \varepsilon_{ep}} \cdot \overset{\triangle}{\varepsilon}_p \\ & + \varrho_R \frac{\partial w_v}{\partial \varepsilon_{ev}} \cdot \overset{\triangle}{\varepsilon}_v + \varrho_R \frac{\partial w_v}{\partial \varepsilon_{ev}} \cdot (\boldsymbol{L}_v{}^T \varepsilon_{ev} + \varepsilon_{ev} \boldsymbol{L}_v) + \varrho_R \frac{\partial w_p}{\partial \varepsilon_{ep}} \cdot (\boldsymbol{L}_p{}^T \varepsilon_{ep} + \varepsilon_{ep} \boldsymbol{L}_p). \end{aligned} \tag{27}$$

Therewith, we can specify the constitutive relations for the overstress $\boldsymbol{\tau}_{ov}$ and the hysteretic part $\boldsymbol{\tau}_{eqp}$ of the equilibrium stress. For physical reasons, we define them by the potential relations

$$\boldsymbol{\tau}_{eqp}(\varepsilon_{ep}, \theta) = \varrho_R \frac{\partial w_p}{\partial \varepsilon_{ep}}, \qquad \tilde{\boldsymbol{T}}_{eqp} = \boldsymbol{F}_p{}^{-1} \boldsymbol{\tau}_{eqp} \boldsymbol{F}_p^{T-1}, \tag{28}$$

$$\boldsymbol{\tau}_{ov}(\varepsilon_{ev}, \theta) = \varrho_R \frac{\partial w_v}{\partial \varepsilon_{ev}}, \qquad \tilde{\boldsymbol{T}}_{ov} = \boldsymbol{F}_v{}^{-1} \boldsymbol{\tau}_{ov} \boldsymbol{F}_v^{T-1} \tag{29}$$

which are completely related to the intermediate configurations $\mathscr{F}_p$ and $\mathscr{F}_v$. The corresponding stress tensors of second Piola Kirchhoff type $\tilde{\boldsymbol{T}}_{ov}$ and $\tilde{\boldsymbol{T}}_{eqp}$ are obtained by the pull-back transformations, specified in Eqs. (28) and (29) as well. The remaining terms in Eq. (27), which contain the inelastic deformation rates $\boldsymbol{L}_v$ and $\boldsymbol{L}_p$ can be simplified if the potentials w_v and w_p are assumed to be isotropic functions of their arguments ε_{ev} and ε_{ep}:

$$\frac{\partial w_v}{\partial \varepsilon_{ev}} \cdot (\boldsymbol{L}_v{}^T \varepsilon_{ev} + \varepsilon_{ev} \boldsymbol{L}_v) = 2\varepsilon_{ev} \frac{\partial w_v}{\partial \varepsilon_{ev}} \cdot \overset{\triangle}{\varepsilon}_v, \qquad \frac{\partial w_p}{\partial \varepsilon_{ep}} \cdot (\boldsymbol{L}_p{}^T \varepsilon_{ep} + \varepsilon_{ep} \boldsymbol{L}_p) = 2\varepsilon_{ep} \frac{\partial w_p}{\partial \varepsilon_{ep}} \cdot \overset{\triangle}{\varepsilon}_p.$$

Inserting these expressions into Eq. (27), the specific entropy production reads

$$\varrho_R \gamma = \varrho_R (1 + 2\varepsilon_{ev}) \frac{\partial w_v}{\partial \varepsilon_{ev}} \cdot \overset{\triangle}{\varepsilon}_v + \varrho_R (1 + 2\varepsilon_{ep}) \frac{\partial w_p}{\partial \varepsilon_{ep}} \cdot \overset{\triangle}{\varepsilon}_p - \frac{\boldsymbol{q}_R \cdot \boldsymbol{g}_R}{\theta}. \tag{30}$$

In order to state sufficient conditions for the non-negativity of dissipation, we specify the evolution equations for the inelastic strains by the following expressions:

$$\overset{\triangle}{\varepsilon}_v := \frac{\varrho_R}{\eta_v} (1 + 2\varepsilon_{ev}) \frac{\partial w_v}{\partial \varepsilon_{ev}}, \qquad \eta_v = \eta_v(\varepsilon_v, \tau_{ov}, \theta) > 0, \tag{31}$$

$$\overset{\triangle}{\varepsilon}_p := \dot{z} \frac{\varrho_R}{\eta_p} (1 + 2\varepsilon_{ep}) \frac{\partial w_p}{\partial \varepsilon_{ep}}, \qquad \eta_p = \eta_p(\varepsilon_p, \tau_{eqp}, \theta) > 0. \tag{32}$$

For physical reasons, the material functions η_v and η_p may depend on the arguments ε_v and τ_{ov} or on ε_p and τ_{eqp} as well as on temperature θ. The quantity $\dot{z}$ in Eq. (32) is the time rate of a kinematic arclength, so that the plastic strain ε_p depends in a rate independent manner on the history of the total deformation. In contrast to classical theories of elasto-plasticity, no yield surface is introduced. For the heat flux $\boldsymbol{q}_R$ we assume the linear Fourier model

$$\boldsymbol{q}_R := -\lambda \boldsymbol{g}_R, \tag{33}$$

where the parameter λ is the coefficient of thermal conductivity. As an essential result of this discussion, we have developed a three-dimensional constitutive theory, which satisfies the dissipation principle for arbitrary temperature-and deformation-processes. Finally, the entropy production is given by the relation

$$\varrho_R \gamma = \frac{1}{\eta_v} \left(\varrho_R (1 + 2\varepsilon_{ev}) \frac{\partial w_v}{\partial \varepsilon_{ev}} \right)^2 + \dot{z} \frac{1}{\eta_p} \left(\varrho_R (1 + 2\varepsilon_{ep}) \frac{\partial w_p}{\partial \varepsilon_{ep}} \right)^2 + \lambda \frac{\boldsymbol{g}_R \cdot \boldsymbol{g}_R}{\theta}, \tag{34}$$

which has exactly the same structure as the corresponding expression for the uniaxial model (Eqs. (7), (8), (9)). Summarising, the three-dimensional theory is defined by the following ingredients: The basic idea comprises two multiplicative decompositions of the deformation gradient into elastic and inelastic parts. Moreover, two additive strain decompositions are introduced by Eqs. (13) as well as the associated stress decompositions by Eqs. (14). Finally the Helmholtz free energy is specified in Eq. (21), the flow rules for the inelastic strains are defined in Eqs. (31) and (32) and Fourier's law of the heat flux is introduced by Eq. (33).

3.3 On the representation of inelastic bulk and distortional effects

At this point, further properties of the mechanical behaviour of rubber are incorporated into the theory. It is a common practice to assume that in elastomers the volume deformation is purely elastic or even zero. In fact, experimental investigations, carried out by Holownia and James [14], show the dynamic bulk modulus of polyurethane rubber to depend only weakly on the frequency of the deformation process. The bulk modulus is about $2{,}5 \times 10^3$ MPa, which is roughly three orders of magnitude larger than the shear stiffness [15], [29], so that under normal operating conditions even the approximation of incompressibility seems to be valid.

Now we apply a concept, which allows a separate modelization of inelastic bulk and distortional effects. In the developed theory these phenomena are related to the overstress and to the hysteretic part of the equilibrium stress.

In order to realise this concept, we consider the deformation gradient and introduce so-called kinematic decompositions: The elastic parts $\boldsymbol{F}_{ev}$ and $\boldsymbol{F}_{ep}$ which are the driving forces for the inelastic stresses are multiplicatively decomposed into a pure volumetric and a distortional part. This idea was proposed by Flory [16], applied to finite viscoelasticity by Lubliner [4] and to finite elasticity by Miehe [23]. Thus, we introduce the following definitions:

$$\boldsymbol{F}_{ev} =: \bar{\boldsymbol{F}}_{ev}\hat{\boldsymbol{F}}_{ev}, \quad J_{ev} := \det(\boldsymbol{F}_{ev}), \quad \bar{\boldsymbol{F}}_{ev} = J_{ev}^{1/3}\,\mathbf{1}, \quad \hat{\boldsymbol{F}}_{ev} = J_{ev}^{-1/3}\,\boldsymbol{F}_{ev} \tag{35.1}$$

$$\boldsymbol{F}_{ep} =: \bar{\boldsymbol{F}}_{ep}\hat{\boldsymbol{F}}_{ep}, \quad J_{ep} := \det(\boldsymbol{F}_{ep}), \quad \bar{\boldsymbol{F}}_{ep} = J_{ep}^{1/3}\,\mathbf{1}, \quad \hat{\boldsymbol{F}}_{ep} = J_{ep}^{-1/3}\,\boldsymbol{F}_{ep}. \tag{35.2}$$

In Eqs. (35.1) and (35.2), the tensors $\bar{\boldsymbol{F}}_{ev}$ and $\bar{\boldsymbol{F}}_{ep}$ denote the volumetric parts of $\boldsymbol{F}_{ev}$ and $\boldsymbol{F}_{ep}$. The unimodular tensors $\hat{\boldsymbol{F}}_{ev}$ and $\hat{\boldsymbol{F}}_{ep}$ are the distortional parts, representing the isochoric part of the motion, and the determinants of $\boldsymbol{F}_{ev}$ and $\boldsymbol{F}_{ep}$ are labelled as J_{ev} and J_{ep}. By Eqs. (35.3) and (35.4), we define so-called volumetric and distortional strain tensors of Right Cauchy Green type $\bar{\boldsymbol{C}}_{ev}$, $\hat{\boldsymbol{C}}_{ev}$, $\bar{\boldsymbol{C}}_{ep}$, $\hat{\boldsymbol{C}}_{ep}$ and of Green type $\bar{\varepsilon}_{ev}$, $\hat{\varepsilon}_{ev}$, $\bar{\varepsilon}_{ep}$, $\hat{\varepsilon}_{ep}$:

$$\boldsymbol{C}_{ev} := \boldsymbol{F}_{ev}^T\boldsymbol{F}_{ev}, \quad \bar{\boldsymbol{C}}_{ev} := \bar{\boldsymbol{F}}_{ev}^T\bar{\boldsymbol{F}}_{ev}, \quad \hat{\boldsymbol{C}}_{ev} := \hat{\boldsymbol{F}}_{ev}^T\hat{\boldsymbol{F}}_{ev}, \quad \bar{\varepsilon}_{ev} := \frac{1}{2}(\bar{\boldsymbol{C}}_{ev} - \mathbf{1}), \quad \hat{\varepsilon}_{ev} := \frac{1}{2}(\hat{\boldsymbol{C}}_{ev} - \mathbf{1}) \tag{35.3}$$

$$\boldsymbol{C}_{ep} := \boldsymbol{F}_{ep}^T\boldsymbol{F}_{ep}, \quad \bar{\boldsymbol{C}}_{ep} := \bar{\boldsymbol{F}}_{ep}^T\bar{\boldsymbol{F}}_{ep}, \quad \hat{\boldsymbol{C}}_{ep} := \hat{\boldsymbol{F}}_{ep}^T\hat{\boldsymbol{F}}_{ep}, \quad \bar{\varepsilon}_{ep} := \frac{1}{2}(\bar{\boldsymbol{C}}_{ep} - \mathbf{1}), \quad \hat{\varepsilon}_{ep} := \frac{1}{2}(\hat{\boldsymbol{C}}_{ep} - \mathbf{1}) \tag{35.4}$$

In addition, we need the derivatives of $\bar{\varepsilon}_{ev}$, $\hat{\varepsilon}_{ev}$ and $\bar{\varepsilon}_{ep}$, $\hat{\varepsilon}_{ep}$ with respect to the elastic strain tensors ε_{ev} and ε_{ep}:

$$\frac{d\bar{\varepsilon}_{ev}}{d\varepsilon_{ev}} = \frac{1}{3}\,J_{ev}^{2/3}\,\mathbf{1}\otimes\boldsymbol{C}_{ev}^{-1}, \quad \frac{d\hat{\varepsilon}_{ev}}{d\varepsilon_{ev}} = J_{ev}^{-2/3}\left[\overset{4}{\mathbf{1}} - \frac{1}{3}\,\boldsymbol{C}_{ev}\otimes\boldsymbol{C}_{ev}^{-1}\right] \tag{35.5}$$

$$\frac{d\bar{\varepsilon}_{ep}}{d\varepsilon_{ep}} = \frac{1}{3}\,J_{ep}^{2/3}\,\mathbf{1}\otimes\boldsymbol{C}_{ep}^{-1}, \quad \frac{d\hat{\varepsilon}_{ep}}{d\varepsilon_{ep}} = J_{ep}^{-2/3}\left[\overset{4}{\mathbf{1}} - \frac{1}{3}\,\boldsymbol{C}_{ep}\otimes\boldsymbol{C}_{ep}^{-1}\right]. \tag{35.6}$$

By means of these definitions and relations we can modify the arguments of the inelastic potentials w_v and w_p. In particular, we replace the elastic strain tensors ε_{ev} and ε_{ep} by their corresponding kinematic decompositions:

$$w_v(\varepsilon_{ev}, \theta) = w_v(\hat{\varepsilon}_{ev}, \bar{\varepsilon}_{ev}, \theta), \qquad w_p(\varepsilon_{ep}, \theta) = w_p(\hat{\varepsilon}_{ep}, \bar{\varepsilon}_{ep}, \theta). \tag{36}$$

In the following part of this discussion we investigate the consequence of this modification on both the inelastic stresses and the flow rules for the inelastic strains. For that, we insert the right hand sides of Eqs. (36) in the potential relations (Eqs. (28), (29)) and apply the chain rule. Bearing Eqs. (35.5) and (35.6) in mind, we obtain the following expressions for the overstress τ_{ov} and the hysteretic part τ_{eqp} of the equilibrium stress:

$$\tau_{ov} = \varrho_R\left(\left[\frac{1}{3}\,J_{ev}^{2/3}\,\frac{\partial w_v}{\partial\bar{\varepsilon}_{ev}}\cdot\mathbf{1}\right]\boldsymbol{C}_{ev}^{-1} + J_{ev}^{-2/3}\left[\frac{\partial w_v}{\partial\hat{\varepsilon}_{ev}} - \frac{1}{3}\,\boldsymbol{C}_{ev}\cdot\frac{\partial w_v}{\partial\hat{\varepsilon}_{ev}}\,\boldsymbol{C}_{ev}^{-1}\right]\right), \tag{37.1}$$

$$\tau_{eqp} = \varrho_R\left(\left[\frac{1}{3}\,J_{ep}^{2/3}\,\frac{\partial w_p}{\partial\bar{\varepsilon}_{ep}}\cdot\mathbf{1}\right]\boldsymbol{C}_{ep}^{-1} + J_{ep}^{-2/3}\left[\frac{\partial w_p}{\partial\hat{\varepsilon}_{ep}} - \frac{1}{3}\,\boldsymbol{C}_{ep}\cdot\frac{\partial w_v}{\partial\hat{\varepsilon}_{ep}}\,\boldsymbol{C}_{ep}^{-1}\right]\right). \tag{37.2}$$

In order to interpret these formulae physically, we compute the related stress tensors of weighted Cauchy type S_{ov} and S_{eqp}, which operate on the current configuration. To this end, we apply the push-forward transformations

$$S_{ov} = F\tilde{T}_{ov}F^T = FF_v^{-1}\tau_{ov}F_v^{T-1}F^T \quad \text{and} \quad S_{eqp} = F\tilde{T}_{eqp}F^T = FF_p^{-1}\tau_{eqp}F_p^{T-1}F^T, \tag{38}$$

and derive the expressions

$$S_{ov} = \varrho_R\left(\left[\frac{1}{3}\frac{\partial w_v}{\partial \bar{\varepsilon}_{ev}}\cdot \bar{C}_{ev}\right]\mathbf{1} + \left[\hat{F}_{ev}\frac{\partial w_v}{\partial \hat{\varepsilon}_{ev}}\hat{F}_{ev}^T - \frac{1}{3}\hat{C}_{ev}\cdot\frac{\partial w_v}{\partial \hat{\varepsilon}_{ev}}\mathbf{1}\right]\right), \tag{39.1}$$

$$S_{eqp} = \varrho_R\left(\left[\frac{1}{3}\frac{\partial w_p}{\partial \bar{\varepsilon}_{ep}}\cdot \bar{C}_{ep}\right]\mathbf{1} + \left[\hat{F}_{ep}\frac{\partial w_p}{\partial \hat{\varepsilon}_{ep}}\hat{F}_{ep}^T - \frac{1}{3}\hat{C}_{ep}\cdot\frac{\partial w_p}{\partial \hat{\varepsilon}_{ep}}\mathbf{1}\right]\right). \tag{39.2}$$

Equations (39) demonstrate that the weighted Cauchy stresses S_{ov} and S_{eqp} split additively into deviatoric and cubic parts. If we assume, for example, the strain energy function w_v to be decomposed into a pure volumetric and a pure distortional, i.e. $w_v(\hat{\varepsilon}_{ev}, \bar{\varepsilon}_{ev}, \theta) = \bar{w}_v(\bar{\varepsilon}_{ev}, \theta) + \hat{w}_v(\hat{\varepsilon}_{ev}, \theta)$, the deviatoric part of the overstress depends only on the distortional part $\hat{\varepsilon}_{ev}$ of strain and the spherical part only on the volumetric part $\bar{\varepsilon}_{ev}$.

In order to investigate the effect of the kinematic decompositions on the evolution equations for the inelastic strains, we insert Eqs. (37) into the flow rules (Eqs. (31) and (32)) and obtain the following formulae:

$$\overset{\triangle}{\varepsilon}_v = \frac{\varrho_R}{\eta_v}\left(\left[\frac{1}{3}\frac{\partial w_v}{\partial \bar{\varepsilon}_{ev}}\cdot \bar{C}_{ev}\right]\mathbf{1} + \left[\hat{C}_{ev}\frac{\partial w_v}{\partial \hat{\varepsilon}_{ev}} - \frac{1}{3}\hat{C}_{ev}\cdot\frac{\partial w_v}{\partial \hat{\varepsilon}_{ev}}\mathbf{1}\right]\right), \tag{40.1}$$

$$\overset{\triangle}{\varepsilon}_p = \dot{z}\,\frac{\varrho_R}{\eta_p}\left(\left[\frac{1}{3}\frac{\partial w_p}{\partial \bar{\varepsilon}_{ep}}\cdot \bar{C}_{ep}\right]\mathbf{1} + \left[\hat{C}_{ep}\frac{\partial w_p}{\partial \hat{\varepsilon}_{ep}} - \frac{1}{3}\hat{C}_{ep}\cdot\frac{\partial w_v}{\partial \hat{\varepsilon}_{ep}}\mathbf{1}\right]\right). \tag{40.2}$$

These equations show that the Oldroyd rates of the inelastic strains decompose additively into deviatoric and spherical parts too. Altogether, we have utilised two different types of strain decompositions in the theory: The first one decomposes the total deformation gradient F multiplicatively into elastic and inelastic parts and is related to the physical properties of the material. This decomposition in combination with the concept of dual variables [7] implies additive strain decompositions with respect to intermediate configurations. The second decomposition is a multiplicative split of the elastic parts F_{ev} and F_{ep} into volumetric and distortional parts and is attributed to the geometry of deformation. In particular, this decomposition implies additive decompositions of the inelastic Cauchy stresses and the inelastic strain rates into spherical and deviatoric parts.

4 Transformation to the reference configuration

In this Section, we prepare the numerical evaluation of the constitutive model. The elastic part of the equilibrium stress operates on the reference configuration (Eq. (24)), whereas the inelastic stresses and the flow rules for the inelastic strains are related to the intermediate configurations $\mathscr{F}_v$ and $\mathscr{F}_p$ (Eqs. (28), (29), (31) and (32)). In order to integrate the corresponding differential equations numerically we have to express the Oldroyd derivatives in terms of ordinary time rates. Accordingly, we transform the total set of constitutive equations to the reference configuration $\mathscr{R}$. The stress tensors, operating on this configuration are of second Piola Kirchhoff type, and the associated strain tensors are of Right Cauchy Green type. This pull-back transformation is the objective of the following discussion.

In the course of the investigation of the thermodynamical properties, the potentials w_v and w_p of the inelastic stress tensors are required to be isotropic functions of their associated elastic strain tensors (Eqs. (28), (29) and (30)), i.e. for example $w_v(\varepsilon_{ev}, \theta) = w_v(I_{ev}, II_{ev}, III_{ev}, \theta)$. In this case I_{ev}, II_{ev} and III_{ev} denote the three invariants of the elastic Right Cauchy Green tensor $\boldsymbol{C}_{ev} = \boldsymbol{F}_{ev}^T\boldsymbol{F}_{ev}$. In order to combine this requirement with the kinematic strain decomposition introduced by Eqs. (36), we propose the following representation formulae for the inelastic strain energies:

$$w_v(\hat{\varepsilon}_{ev}, \bar{\varepsilon}_{ev}, \theta) = w_v(\hat{\boldsymbol{C}}_{ev}, \bar{\boldsymbol{C}}_{ev}, \theta) = w_v(\hat{I}_{ev}, \hat{II}_{ev}, \overline{III}_{ev}, \theta), \tag{41.1}$$

$$w_p(\hat{\varepsilon}_{ep}, \bar{\varepsilon}_{ep}, \theta) = w_p(\hat{\boldsymbol{C}}_{ep}, \bar{\boldsymbol{C}}_{ep}, \theta) = w_p(\hat{I}_{ep}, \hat{II}_{ep}, \overline{III}_{ep}, \theta). \tag{41.2}$$

In Eqs. (41), the variables $\hat{I}_{ev}$, $\hat{II}_{ev}$, and $\hat{I}_{ep}$, $\hat{II}_{ep}$ denote the first and second invariants of the distortional elastic Right Cauchy Green tensors $\hat{\boldsymbol{C}}_{ev}$ and $\hat{\boldsymbol{C}}_{ep}$ defined by Eqs. (35), whereas $\overline{III}_{ev}$ and $\overline{III}_{ep}$ are the determinants of the volumetric tensors $\bar{\boldsymbol{C}}_{ev}$ and $\bar{\boldsymbol{C}}_{ep}$. The following set of formulae (Eqs. (42) to (45)) demonstrates, that all invariants which occur in Eqs. (41) can be expressed in terms of the deformation tensors $\boldsymbol{C} = \boldsymbol{F}^T\boldsymbol{F}$, $\boldsymbol{C}_v$, and $\boldsymbol{C}_p$:

$$\boldsymbol{C}_v := \boldsymbol{F}_v{}^T\boldsymbol{F}_v, \quad \boldsymbol{C}_p := \boldsymbol{F}_p{}^T\boldsymbol{F}_p, \quad \frac{1}{2}\dot{\boldsymbol{C}}_v = \boldsymbol{F}_v{}^T\overset{\triangle}{\varepsilon}_v\boldsymbol{F}_v, \quad \frac{1}{2}\dot{\boldsymbol{C}}_p = \boldsymbol{F}_p{}^T\overset{\triangle}{\varepsilon}_p\boldsymbol{F}, \tag{42}$$

$$\overline{III}_{ev} = \det \bar{\boldsymbol{C}}_{ev} = \frac{\det \boldsymbol{C}}{\det \boldsymbol{C}_v}, \quad \overline{III}_{ep} = \det \bar{\boldsymbol{C}}_{ep} = \frac{\det \boldsymbol{C}}{\det \boldsymbol{C}_p}, \tag{43}$$

$$\hat{I}_{ev} = tr\hat{\boldsymbol{C}}_{ev} = \left(\frac{\det \boldsymbol{C}_v}{\det \boldsymbol{C}}\right)^{1/3} \boldsymbol{C} \cdot \boldsymbol{C}_v{}^{-1}, \quad \hat{I}_{ep} = tr\hat{\boldsymbol{C}}_{ep} = \left(\frac{\det \boldsymbol{C}_p}{\det \boldsymbol{C}}\right)^{1/3} \boldsymbol{C} \cdot \boldsymbol{C}_p{}^{-1}, \tag{44}$$

$$\hat{II}_{ev} = \frac{(tr\hat{\boldsymbol{C}}_{ev})^2 - tr\hat{\boldsymbol{C}}_{ev}^2}{2} = \left(\frac{\det \boldsymbol{C}_v}{\det \boldsymbol{C}}\right)^{2/3} \frac{(tr\boldsymbol{C}\boldsymbol{C}_v{}^{-1})^2 - tr(\boldsymbol{C}\boldsymbol{C}_v{}^{-1})^2}{2}, \tag{45.1}$$

$$\hat{II}_{ep} = \frac{(tr\hat{\boldsymbol{C}}_{ep})^2 - tr\hat{\boldsymbol{C}}_{ep}^2}{2} = \left(\frac{\det \boldsymbol{C}_p}{\det \boldsymbol{C}}\right)^{2/3} \frac{(tr\boldsymbol{C}\boldsymbol{C}_p{}^{-1})^2 - tr(\boldsymbol{C}\boldsymbol{C}_p{}^{-1})^2}{2}. \tag{45.2}$$

In order relate the overstress $\boldsymbol{\tau}_{ov}$ and the hysteretic part of the equilibrium stress $\boldsymbol{\tau}_{eqp}$ to the reference configuration, we apply the pull-back transformations, specified in Eqs. (28) and (29), to Eqs. (37) and utilise Eqs. (42) to (45). After elementary calculations, we obtain the following expressions for the inelastic stresses of Piola Kirchhoff type:

$$\begin{aligned}\tilde{\boldsymbol{T}}_{ov} = \frac{2}{\varrho_R}\Bigg(&\left[\overline{III}_{ev}\frac{\partial w_v}{\partial \overline{III}_{ev}} - \frac{1}{3}\left[\hat{I}_{ev}\frac{\partial w_v}{\partial \hat{I}_{ev}} + 2\hat{II}_{ev}\frac{\partial w_v}{\partial \hat{II}_{ev}}\right]\right]\boldsymbol{C}^{-1} \\ &+ \frac{1}{\overline{III}_{ev}^{1/3}}\left[\frac{\partial w_v}{\partial \hat{I}_{ev}} + \hat{I}_{ev}\frac{\partial w_v}{\partial \hat{II}_{ev}}\right]\boldsymbol{C}_v{}^{-1} - \frac{1}{\overline{III}_{ev}^{2/3}}\frac{\partial w_v}{\partial \hat{II}_{ev}}\boldsymbol{C}_v{}^{-1}\boldsymbol{C}\boldsymbol{C}_v{}^{-1}\Bigg),\end{aligned} \tag{46.1}$$

$$\begin{aligned}\tilde{\boldsymbol{T}}_{eqp} = \frac{2}{\varrho_R}\Bigg(&\left[\overline{III}_{ep}\frac{\partial w_p}{\partial \overline{III}_{ep}} - \frac{1}{3}\left[\hat{I}_{ep}\frac{\partial w_p}{\partial \hat{I}_{ep}} + 2\hat{II}_{ep}\frac{\partial w_p}{\partial \hat{II}_{ep}}\right]\right]\boldsymbol{C}^{-1} \\ &+ \frac{1}{\overline{III}_{ep}^{1/3}}\left[\frac{\partial w_p}{\partial \hat{I}_{ep}} + \hat{I}_{ep}\frac{\partial w_p}{\partial \hat{II}_{ep}}\right]\boldsymbol{C}_p{}^{-1} - \frac{1}{\overline{III}_{ep}^{2/3}}\frac{\partial w_p}{\partial \hat{II}_{ep}}\boldsymbol{C}_p{}^{-1}\boldsymbol{C}\boldsymbol{C}_p{}^{-1}\Bigg).\end{aligned} \tag{46.2}$$

Considering the pull-back transformations for the deformation rate tensors in Eq. (42) and using Eqs. (43) to (45) we can transform the flow rules for the inelastic strains, specified in Eqs. (40), to the reference configuration. Finally, we obtain the following expressions for the time rates of the so-called inelastic Right Cauchy Green tensors:

$$\dot{\boldsymbol{C}}_v = \frac{4\varrho_R}{\eta_v}\left(\left[\hat{III}_{ev}\frac{\partial w_v}{\partial \hat{III}_{ev}} - \frac{1}{3}\left[\hat{I}_{ev}\frac{\partial w_v}{\partial \hat{I}_{ev}} + 2\hat{II}_{ev}\frac{\partial w_v}{\partial \hat{II}_{ev}}\right]\right]\boldsymbol{C}_v\right.$$
$$\left. + \frac{1}{\hat{III}_{ev}^{1/3}}\left[\frac{\partial w_v}{\partial \hat{I}_{ev}} + \hat{I}_{ev}\frac{\partial w_v}{\partial \hat{II}_{ev}}\right]\boldsymbol{C} - \frac{1}{\hat{III}_{ev}^{2/3}}\frac{\partial w_v}{\partial \hat{II}_{ev}}\boldsymbol{C}\boldsymbol{C}_v^{-1}\boldsymbol{C}\right), \tag{47.1}$$

$$\dot{\boldsymbol{C}}_v = \dot{z}\,\frac{4\varrho_R}{\eta_p}\left(\left[\hat{III}_{ep}\frac{\partial w_p}{\partial \hat{III}_{ep}} - \frac{1}{3}\left[\hat{I}_{ep}\frac{\partial w_p}{\partial \hat{I}_{ep}} + 2\hat{II}_{ep}\frac{\partial w_p}{\partial \hat{II}_{ep}}\right]\right]\boldsymbol{C}_p\right.$$
$$\left. + \frac{1}{\hat{III}_{ep}^{1/3}}\left[\frac{\partial w_p}{\partial \hat{I}_{ep}} + \hat{I}_{ep}\frac{\partial w_p}{\partial \hat{II}_{ep}}\right]\boldsymbol{C} - \frac{1}{\hat{III}_{ep}^{2/3}}\frac{\partial w_p}{\partial \hat{II}_{ep}}\boldsymbol{C}\boldsymbol{C}_p^{-1}\boldsymbol{C}\right). \tag{47.2}$$

For the sake of completeness, we specify now the elastic part $\tilde{\boldsymbol{T}}_{eqo}$ of the equilibrium stress. It should be remarked that the associated strain energy function w_o needs not necessarily be isotropic. In the case of isotropy, we propose the representation

$$w_0(\boldsymbol{E}, \theta) = w_0(\boldsymbol{C}, \theta) = w_0(I_C, II_C, III_C, \theta), \tag{48}$$

so that the corresponding stress tensor $\tilde{\boldsymbol{T}}_{eqo}$ is given by the expression

$$\tilde{\boldsymbol{T}}_{eqo} = 2\varrho_R\left(\left[\frac{\partial w_0}{\partial I_C} + I_C\frac{\partial w_0}{\partial II_C}\right]\boldsymbol{1} - \frac{\partial w_0}{\partial II_C}\boldsymbol{C} + \left[III_C\frac{\partial w_0}{\partial III_C}\right]\boldsymbol{C}^{-1}\right). \tag{49}$$

At this point, the pull-back transformation of the constitutive model is complete. This discussion has shown, that the total set of constitutive equations can easily be related to the reference configuration although the development of the theory was based on three different configurations. Summarising the result, the total set of equations with respect to the reference configuration is defined by the additive stress decomposition in Eq. (14), the potential relations in Eqs. (46) and (49), the potentials in Eqs. (41) and (48), and finally the differential equations for the inelastic strains which are specified in Eqs. (47).

4 Experimental observations and numerical simulations

In order to rate the quality of the proposed constitutive theory, experimental observations on cylindrical specimens of carbon black filled rubber (total length: 67 mm, initial length of the measuring section: 20 mm, initial diameter: 20 mm) are discussed and compared with numerical simulations. The investigated material was a rather soft tread compound, provided by the Continental AG in Hannover (Germany). A detailed description of the experimental results which are shown in this paper can be found in [1], [31]. All tests were performed at room temperature under strain control. The elongation of the measuring section was determined by a laser extensometer, and the engineering strain was calculated under the assumption of homogeneous deformations. The strain rate is the rate of engineering strain, and the engineering stress is the axial force per cross-sectional area of the undeformed specimen.

If mechanical loads are applied, filler loaded rubber shows a certain kind of softening behaviour which is known in the literature as the Mullins effect. One basic problem which arises in the investigation of the mechanical properties of such materials is the separation of this effect from the other phenomena of interest. Phenomenologically, the Mullins effect can be interpreted as a damage phenomenon which occurs essentially during the first periods of the loading process. A detailed description of this effect is given for example by James and Green [15] or by Johnson and Beatty [17], [18]. In order to separate the Mullins effect from the mechanical phenomena which are in the centre of interest of this work, we have treated all virgin specimen by a mechanical pre-process until a stable state of the material is reached (see for example [1], [15]). After this pre-process, the actual experiments can be carried out. The pre-process applied in [1] consists of 12 loading cycles under strain control. The strain range was 0.3 in compression and 1.0 in tension and the absolute value of the strain rate was about $2 \times 10^{-2} s^{-1}$. Subsequent to this, a relaxation period of 1 hour at zero strain was introduced. Experimentally, it is observed that during the first cycles of the pre-process softening occurs just until a stationary stress amplitude is reached. In this situation, the material is in a stable state. At the end of the relaxation period, the stress is observed to be approximately zero. The corresponding experimental data sets are shown in [1].

Figure 3 shows a set of 10 monotonic experiments under strain control in tension and in compression, carried out just after the pre-process. Each test was run using a new specimen. The first 8 experiments correspond to the following values of the strain rate: $\pm 2 \times 10^{-4} s^{-1}$, $\pm 2 \times 10^{-3} s^{-1}$, $\pm 2 \times 10^{-2} s^{-1}$, $\pm 2 \times 10^{-1} s^{-1}$, whereas in two further experiments the constant strain rate was interrupted by a series of hold times at constant strain with a duration of 1 hour. A comparison between the curves in Fig. 3 shows that in tension as well as in compression the stress increases nonlinearly with the applied strain rate. In addition, it is observed that relaxation occurs during the hold times. If the duration of the hold times is long enough, then so-called time independent equilibrium states are approximately reached. The connecting curve of these equilibrium states is defined as the *equilibrium stress strain curve* and the difference between the total stress and the equilibrium stress is the *overstress*. In [1], [6] the physical justification of these definitions is discussed.

By means of the tests in Figs. 4 and 5, the material behaviour under cyclic loading conditions is investigated. Figure 4 shows three experiments with a strain amplitude of 0.3 and various values of mean strain. The absolute value of the strain rate was about $2 \times 10^{-2} s^{-1}$. Subsequent to the pre-process, the strain was increased up to the mean strain. In the first experiment the mean strain was zero, in the second it was about 0.35 and in the last test about 0.7. Then 13 cycles with a constant strain amplitude were applied. In order to investigate the history dependence of the equilibrium states, the last cycles were interrupted by a series of relaxation periods at constant strain. The duration was about 1 hour as before. Figure 5 shows a set of corresponding tests with a strain amplitude of 0.15, where the values of mean strain were about -0.15, 0.35 and 0.85. Summarising, Figs. 4 and 5 show that the general shape of the stress strain response behaviour of carbon black reinforced rubber is mainly influenced by *nonlinear elastic* behaviour. Furthermore, certain *hysteresis effects* occur. A comparison between the asymptotic stress values reached at the termination points of the relaxation periods indicates, that the equilibrium stress response seems to be accompanied by weak hysteresis effects as well. This is the so-called *equilibrium hysteresis.*

Altogether, the experimental results have shown that the stabilised mechanical behaviour of carbon black filled rubber at room temperature is mainly influenced by elasticity coupled with nonlinear rate dependence and weak equilibrium hysteresis effects.

The discussion of the experimental data has now been completed and the observations will be compared with the numerical simulations, based on the theory. The starting point for the

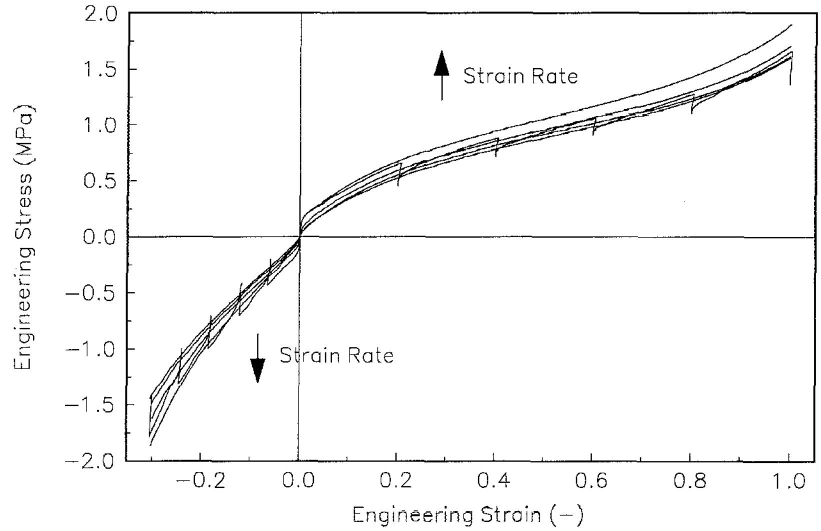

Fig. 3. Monotonic experiments with different strain rates in tension and compression ($\pm 2 \times 10^{-4} s^{-1}$, $\pm 2 \times 10^{-3} s^{-1}$, $\pm 2 \times 10^{-2} s^{-1}$, $\pm 2 \times 10^{-1} s^{-1}$)

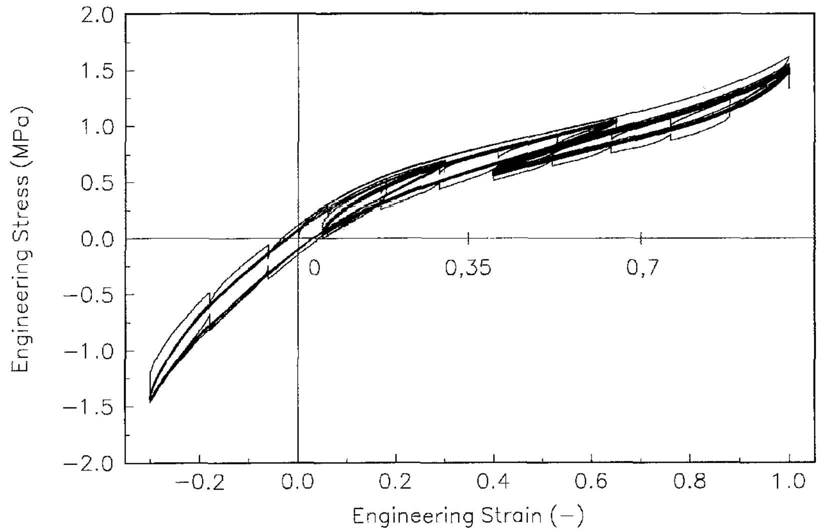

Fig. 4. Cyclic experiments with a strain amplitude of 0.3 and different values of mean strain (0.0, 0.35, 0.7)

development of the constitutive theory was a rheological model, which incorporates the main ideas. Then, the structure of the model was generalised to the threedimensional case. Within the developed model, all experimentally observed phenomena are incorporated: Nonlinear elasticity, nonlinear rate dependence and equilibrium hysteresis effects.

In order to illustrate this ability, the three-dimensional model represented by Eqs. (46), (47) and (49), is numerically integrated for the same set of uniaxial isothermal monotonic and cyclic loading processes in tension and in compression as applied in the experiments. For simplification we make some special assumptions. At first we introduce the approximation of incompressibility, namely

$$\det \boldsymbol{F} = \det \boldsymbol{F}_v = \det \boldsymbol{F}_{ev} = \det \boldsymbol{F}_p = \det \boldsymbol{F}_{ep} = 1, \tag{50.1}$$

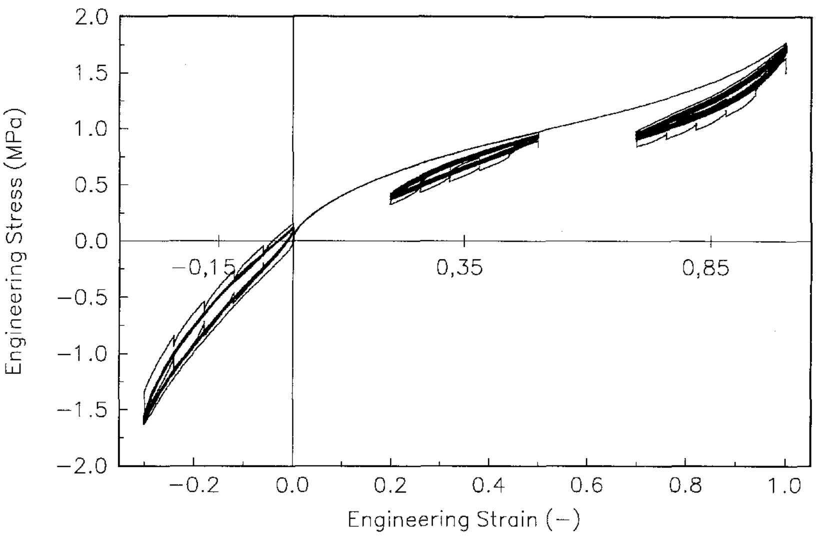

Fig. 5. Cyclic experiments with a strain amplitude of 0.15 and different values of mean strain (−0.15, 0.35, 0.85)

so that the corresponding deformation gradient has the representation

$$F = \begin{bmatrix} 1+\varepsilon & 0 & 0 \\ 0 & \dfrac{1}{\sqrt{1+\varepsilon}} & 0 \\ 0 & 0 & \dfrac{1}{\sqrt{1+\varepsilon}} \end{bmatrix} \tag{50.2}$$

In Eq. (50.2) the variable ε denotes the axial strain. In this case, the second Piola Kirchhoff stress is given by

$$\tilde{T} = -pC^{-1} + \tilde{T}_{eqo} + \tilde{T}_{eqp} + \tilde{T}_{ov}, \tag{51}$$

where the pressure p is not determined by the history of the deformation but by the balance of momentum. The next step is the identification of the material functions and parameters. These follow from a lot of experience in combination with a large number of numerical simulations and comparisons with the experimental data. In particular the potential function w_0, which is attributed to the entropic thermoelastic behaviour of the material, is specialised to be of a modified Mooney-Rivlin type (see also Miehe [25] and Holzapfel [29]):

$$\varrho_R w_0(E, \theta) = \frac{\theta}{\theta_0}\left(c_{o1}(I_C - 3) + c_{o2}(II_C - 3) + c_{o3}(I_C - 3)^5\right) \tag{52}$$

The material parameters are given by $c_{o1} = 0.7\ MPa$, $c_{o2} = 0.1\ MPa$ and $c_{o3} = 0.002\ MPa$, whereas θ_o denotes a given reference temperature (for example room temperature: $\theta_o = 296\ K$). The nonlinear term in Eq. (52) is introduced to model the progressive increase of stress at large values of the strain. The potentials w_p and w_v of the inelastic stresses which correspond to

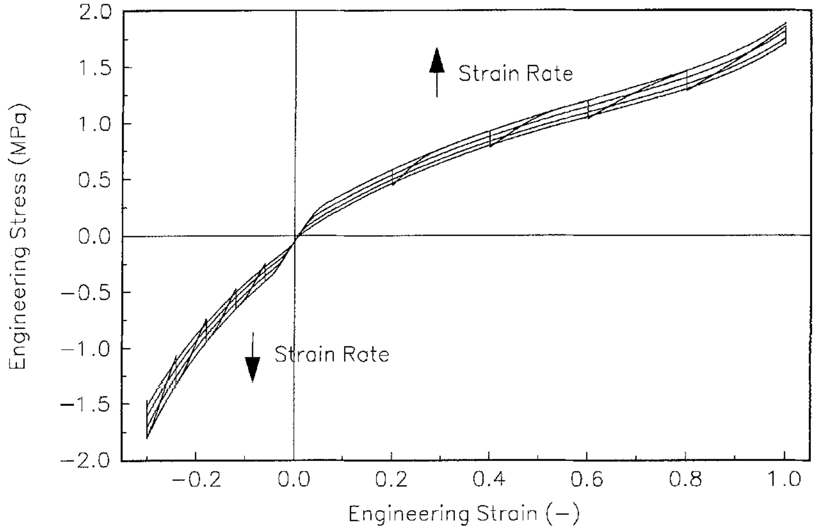

Fig. 6. Monotonic simulations with different strain rates in tension and compression ($\pm 2 \times 10^{-4}s^{-1}$, $\pm 2 \times 10^{-3}s^{-1}$, $\pm 2 \times 10^{-2}s^{-1}$, $\pm 2 \times 10^{-1}s^{-1}$)

the hysteresis effects are expressed as

$$\varrho_R w_p(\varepsilon_{ep}, \theta) = c_{p1}(\hat{I}_{ep} - 3) + c_{p2}(\hat{II}_{ep} - 3), \tag{53}$$

$$\varrho_R w_v(\varepsilon_{ev}, \theta) = c_{v2}(\hat{II}_{ev} - 3), \tag{54}$$

where the material parameters are given by $c_{p1} = 0.05$ MPa, $c_{p2} = 0.05$ MPa and $c_{v2} = 0.6$ MPa. The expression for the rate of the kinematic arclegth $z(t)$ in Eq. (47.2) is specified as $\dot{z} = \|\dot{\mathbf{C}}\|$, where $\mathbf{C}$ is the Right Cauchy Green tensor. Finally, the nonlinear viscosity function η_v is represented by the exponential law

$$\eta_v = \eta_{vo}\exp\left[-\frac{\|\boldsymbol{\tau}_{ov}\|}{s_o\|\mathbf{1} + 2\boldsymbol{\varepsilon}_v\|^3}\right], \qquad \|\boldsymbol{\tau}_{ov}\| = \sqrt{\mathrm{tr}(\tilde{\mathbf{T}}_{ov}\mathbf{C}_v)^2}, \qquad \|\mathbf{1} + 2\boldsymbol{\varepsilon}_v\| = \sqrt{\mathrm{tr}(\mathbf{C}_v{}^{-2})}, \tag{55}$$

where the material constant are given by $\eta_v = 100$ Pas and $s_o = 0.02$ MPa. The material function η_p in the evolution law for the plastic strain is assumed to be constant: $\eta_p = 0.25$ MPa.

Now, the preparations are complete and we can carry out the numerical simulations. Because of the history dependence, all simulations were computed subsequent to the 'numerical application' of the pre-process discussed before. A comparison between Figs. 3 and 6 shows that the strongly nonlinear stress strain behaviour under monotonic loadings both in tension and in compression is well described. In addition, the nonlinear rate dependence over four orders of magnitude of the strain rate is reproduced by the theory. A comparison between the cyclic experiments in Figs. 4 and 5 with the corresponding simulations in Figs. 7 and 8 suggests that both the rate dependent hysteresis effects and the equilibrium hysteresis phenomena are represented.

5 A thermomechanical coupling problem

In the previous Section we have shown that the constitutive theory describes the mechanical properties of carbon black reinforced rubber at room temperature very well. As a first step up to the representation of thermomechanical properties, we demonstrate the ability of the model to

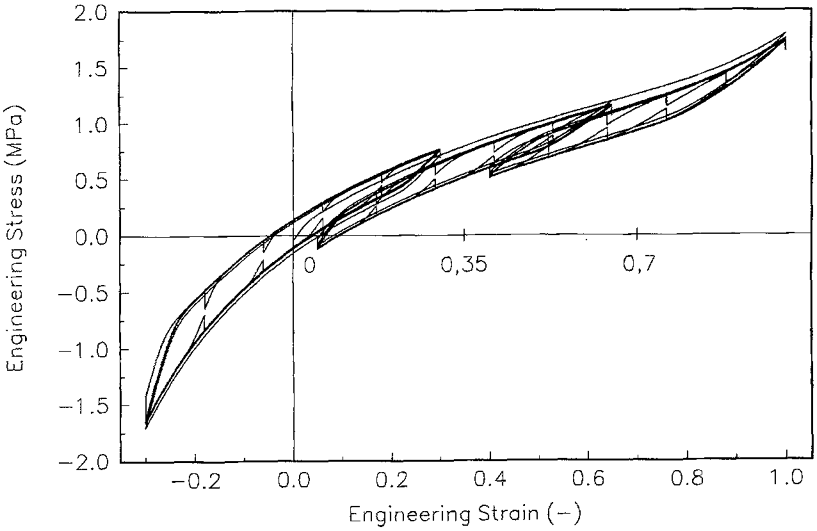

Fig. 7. Cyclic simulations with a strain amplitude of 0.3 and different values of mean strain (0.0, 0.35, 0.7)

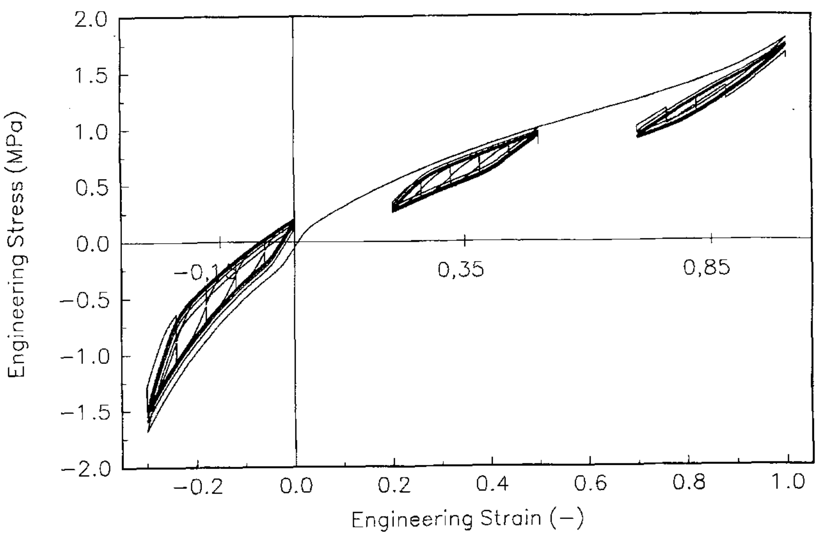

Fig. 8. Cyclic simulations with a strain amplitude of 0.15 and different values of mean strain (−0.15, 0.35, 0.85)

simulate dissipative heating phenomena. In general it is rather complicated to solve thermomechanically coupled boundary value problems: The time-dependent strain field in combination with the inelastic material properties leads to an increase of temperature which influences the material properties and therewith the strain field and so on. Numerical methods to solve such problems are proposed for example by Miehe [23], [26]. For this reason, we discuss a rather simple problem which can be solved analytically: A cylindrical specimen of carbon black filled rubber is loaded by a cyclic displacement controlled process. The displacement amplitude, the mean value, and the loading frequency are prescribed, and the temporal mean values of the stationary temperature distribution are calculated by the theory. For simplification, we make the assumption of temperature independent material parameters, which holds if the temperature increase is small in comparison with the reference temperature (see Kamlah [40] and Kamlah et al. [41]). The numerical simulations are based on the material-functions and -parameters, which are specified in the last Section.

In order to discuss this problem physically correct, we have to derive the equation of heat conduction from the first law of thermodynamics in combination with the constitutive model. The first law of thermodynamics is given by the expression

$$\varrho_R(\psi + \theta s)^{\cdot} = -\mathrm{Div}\boldsymbol{q}_R + \tilde{\boldsymbol{T}} \cdot \dot{\boldsymbol{E}} + \varrho_R r, \tag{56}$$

where the internal energy has been expressed in terms of the Helmholtz free energy ψ, the entropy s and the absolute temperature θ. The variable r denotes the specific heat supply by radiation. In view of Eqs. (21) and (52) to (54) we can specialise the free energy per unit mass as follows:

$$\psi = \theta v_o(\boldsymbol{E}) + w_p(\boldsymbol{\varepsilon}_{ep}) + w_v(\boldsymbol{\varepsilon}_{ev}) + \varphi(\theta). \tag{57}$$

The potentials w_p and w_v are assumed to be independent of temperature [21], and the strain energy function w_o which determines the elastic part of the stress, is postulated to depend linearly on the temperature (see [11], [19], [25], [29]):

$$w_o(\boldsymbol{E}, \theta) = \theta v_o(\boldsymbol{E}). \tag{58}$$

Utilising the potential relation specified in Eq. (25), and the Helmholtz free energy of Eq. (57), we obtain the following formulae for the specific entropy s and its corresponding time rate:

$$s = -[v_o(\boldsymbol{E}) + \varphi'(\theta)] \tag{59}$$

$$\theta \dot{s} = -\theta \frac{\partial v_o}{\partial \boldsymbol{E}} \cdot \dot{\boldsymbol{E}} - c_D \dot{\theta}, \qquad -\theta\varphi''(\theta) =: c_D(\theta) = c_D = \text{const.} \tag{60}$$

In Eq. (60), the coefficient c_D is the specific heat capacity per unit mass (see [20]). For simplification, we assume that c_D is temperature independent. A further expression for the rate of the specific entropy is obtained by the evaluation of the first law of thermodynamics. Inserting the free energy function into Eq. (56), considering the potential relation for the elastic part of the equilibrium stress, Eq. (24), and rearranging terms leads to the expression

$$\varrho_R \theta \dot{s} = -\mathrm{Div}\boldsymbol{q}_R + \tilde{\boldsymbol{T}}_{eqp} \cdot \dot{\boldsymbol{E}} + \tilde{\boldsymbol{T}}_{ov} \cdot \dot{\boldsymbol{E}} - \varrho_R \frac{\partial w_p}{\partial \boldsymbol{\varepsilon}_{ep}} \cdot \dot{\boldsymbol{\varepsilon}}_{ep} - \varrho_R \frac{\partial w_v}{\partial \boldsymbol{\varepsilon}_{ev}} \cdot \dot{\boldsymbol{\varepsilon}}_{ev} + \varrho_R r. \tag{61}$$

Now, we replace the inelastic stress power terms in Eq. (61) by the inner products of the dual stress tensors with their associated strain rates (Eqs. (20)), and the time rates of the elastic strains $\boldsymbol{\varepsilon}_{ev}$ and $\boldsymbol{\varepsilon}_{ep}$ by their corresponding Oldroyd derivatives. After elementary calculations we obtain the following relation:

$$\varrho_R \theta \dot{s} = -\mathrm{Div}\boldsymbol{q}_R + \varrho_R(\boldsymbol{1} + 2\boldsymbol{\varepsilon}_{ev}) \frac{\partial w_p}{\partial \boldsymbol{\varepsilon}_{ev}} \cdot \overset{\triangle}{\boldsymbol{\varepsilon}}_v + \varrho_R(\boldsymbol{1} + 2\boldsymbol{\varepsilon}_{ep}) \frac{\partial w_p}{\partial \boldsymbol{\varepsilon}_{ep}} \cdot \overset{\triangle}{\boldsymbol{\varepsilon}}_p + \varrho_R r. \tag{62}$$

After this, we insert the constitutive equation for the heat flux $\boldsymbol{q}_R$ specified in Eq. (33) into Eq. (62), and replace the entropy rate $\dot{s}$ by Eq. (60). Considering the flow rules for the inelastic strains (Eqs. (31) and (32)), we obtain the so-called three-dimensional equation of heat conduction:

$$\begin{aligned} \frac{\partial \theta}{\partial t} = \frac{\lambda}{\varrho_R c_D} \Delta\theta &+ \frac{\varrho_R}{c_D}\left(\frac{1}{\eta_v(\boldsymbol{\varepsilon}_v, \boldsymbol{\tau}_{ov}, \theta)} \left\| \boldsymbol{C}_{ev} \frac{\partial w_v}{\partial \boldsymbol{\varepsilon}_{ev}} \right\|^2 + \frac{\dot{z}}{\eta_p(\boldsymbol{\varepsilon}_p, \boldsymbol{\tau}_{eqp}, \theta)} \left\| \boldsymbol{C}_{ep} \frac{\partial w_p}{\partial \boldsymbol{\varepsilon}_{ep}} \right\|^2 \right) \\ &+ \frac{r}{c_D} + \frac{\theta}{c_D} \frac{\partial v_o}{\partial \boldsymbol{E}} \cdot \dot{\boldsymbol{E}}. \end{aligned} \tag{63}$$

This is a nonlinear differential equation for the temperature distribution $\theta(X, t)$, where Δ denotes the three-dimensional Laplace operator. The terms in brackets describe the dissipative heat production, caused by inelastic deformations. Eq. (63) shows that the heat production is temperature dependent in general, although the potentials w_v and w_p have been assumed to be temperature independent (Eq. (57)). This temperature dependence is inferred by the material functions η_v and η_p. The term on the right hand side of Eq. (63) is due to the thermoelastic coupling effect.

In the following, we specialise this equation of heat conduction to the uniaxial case and solve it for the problem, which is illustrated in Fig. 9: A cylindrical specimen of rubber is loaded by a cyclic process:

$$u(t) = u_o + \Delta u \sin(\omega t). \tag{64}$$

The variable $u(t)$ denotes the relative displacement, u_o the mean value and Δu the amplitude. The angular frequency is denoted by ω, and the engineering strain is given by $u(t)/l_o$. From now on, we assume that the temperature changes $\vartheta(x, t)$, which are caused by inelastic deformations are small in comparison with the constant room temperature θ_0:

$$\theta(x, t) =: \theta_0 + \vartheta(x, t), \qquad |\vartheta(x, t)| \ll \theta_o.$$

Then, the approximation of the temperature independence of the functions η_v and η_p is valid,

$$\begin{aligned} &\eta_p(\varepsilon_p, \tau_{eqp}, \theta) \approx \eta_p(\varepsilon_p, \tau_{eqp}, \theta_o) =: \eta_p(\varepsilon_p, \tau_{eqp}), \\ &\eta_v(\varepsilon_v, \tau_{ov}, \theta) \approx \eta_v(\varepsilon_v, \tau_{ov}, \theta_o) =: \eta_v(\varepsilon_v, \tau_{ov}), \end{aligned} \tag{65}$$

and the dissipative heat production in Eq. (63) is only a function of time and not of space. Now we suppose that isothermal boundary conditions preside in axial direction and adiabatic boundary conditions in radial direction. In this case, ϑ is a function solely of the axial co-ordinate x and of the time t. Under these assumptions as well as setting the heat supply r at zero, we obtain the uniaxial form of the linearized equation of heat conduction:

$$\begin{aligned} \frac{\partial \vartheta}{\partial t} = \frac{\lambda}{\varrho_R c_D} \frac{\partial^2 \vartheta}{\partial x^2} + \frac{\varrho_R}{c_D} \left(\frac{1}{\eta_v(\varepsilon_v, \tau_{ov})} \left\| \boldsymbol{C}_{ev} \frac{\partial w_v}{\partial \varepsilon_{ev}} \right\|^2 + \frac{\dot{z}}{\eta_p(\varepsilon_p, \tau_{eqp})} \left\| \boldsymbol{C}_{ep} \frac{\partial w_p}{\partial \varepsilon_{ep}} \right\|^2 \right) \\ + \frac{\theta_o}{c_D} \frac{\partial v_o}{\partial \boldsymbol{E}} \cdot \dot{\boldsymbol{E}}, \end{aligned} \tag{66.1}$$

$$\vartheta(0, t) = \vartheta(l_o, t) = 0 \tag{66.2}$$

The corresponding boundary conditions are formulated by Eq. (66.2). In the following, we calculate a stationary solution of Eqs. (66), which belongs to the loading process of Eq. (64).

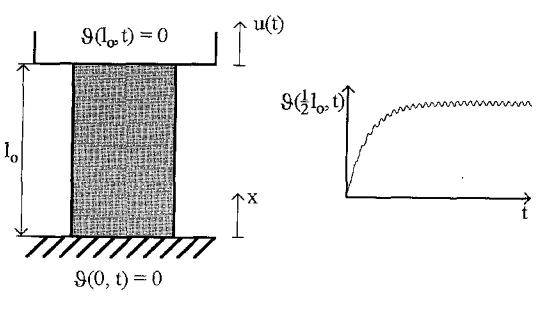

Fig. 9. Thermomechanical coupling problem

In the case of uniaxial cyclic strain controlled loadigs and under the assumption of incompressible material behaviour, the thermoelastic coupling term in Eq. (66.1) is a periodic function of time from the very beginning. On the other hand, the dissipative heat production changes periodically only in the asymptotic limit. The corresponding temperature function is illustrated qualitatively in Fig. 9: Subsequent to a rapid increase, a stationary temperature distribution is reached. This behaviour can be characterised by a time independent mean value, which is superimposed by rather small oscillations. Consequently, the stationary temperature distribution is a periodic function of time, i.e. $\vartheta(x, t) = \vartheta(x, t + T)$, where the period T is determined by the inverse frequency of the loading process $2\pi/\omega$. In order to estimate the stationary temperature distribution, we introduce the temporal mean value $\vartheta_M(x)$ of the temperature function $\vartheta(x, t)$ as follows:

$$\vartheta_M(x) := \frac{1}{T} \int_t^{t+T} \vartheta\,(x, \tau)\, d\tau \quad \Rightarrow \quad \vartheta''_M(x) = \frac{1}{T} \int_t^{t+T} \frac{\partial^2 \vartheta}{\partial x^2}(x, \tau)\, d\tau\,. \tag{67}$$

Integration of Eq. (66.1) over one period with respect to time and utilising the formulae

$$\int_t^{t+T} \frac{\partial \vartheta}{\partial \tau}(x, \tau)\, d\tau = 0, \qquad \frac{\theta_o}{c_D} \int_t^{t+T} \frac{\partial v_o}{\partial \mathbf{E}} \cdot \dot{\mathbf{E}}(\tau)\, d\tau = 0\,, \tag{68}$$

which result from the periodicity of temperature and strain, we obtain an ordinary differential equation for $\vartheta_M(x)$:

$$\vartheta''_M(x) = -\frac{\varrho_R{}^2}{\lambda T} \int_t^{t+T} \left(\frac{1}{\eta_v(\varepsilon_v, \tau_{ov})} \left\| C_{ev} \frac{\partial w_v}{\partial \varepsilon_{ev}} \right\|^2 + \frac{\dot{z}}{\eta_p(\varepsilon_p, \tau_{eqp})} \left\| C_{ep} \frac{\partial w_p}{\partial \varepsilon_{ep}} \right\|^2 \right)(\tau)\, d\tau\,. \tag{69}$$

Equation (69) shows that the temporal mean value of the stationary temperature distribution depends only on the temporal mean value of the dissipation. The solution of Eq. (69) with respect to the boundary conditions in Eq. (66.2) is given by the expression

$$\vartheta_M(x) = \frac{\varrho_R{}^2\, x(l_o - x)}{2\lambda T} \int_t^{t+T} \left(\frac{1}{\eta_v(\varepsilon_v, \tau_{ov})} \left\| C_{ev} \frac{\partial w_v}{\partial \varepsilon_{ev}} \right\|^2 + \frac{\dot{z}}{\eta_p(\varepsilon_p, \tau_{eqp})} \left\| C_{ep} \frac{\partial w_p}{\partial \varepsilon_{ep}} \right\|^2 \right)(\tau)\, d\tau\,. \tag{70}$$

The mean value of the temperature depends quadratically on the co-ordinate x, so that the maximum is reached in the centre of the specimen. By using Eq. (70), we demonstrate now the ability of the theory to simulate dissipative heating phenomena and calculate the temperatures $\vartheta_M(x)$. The temporal mean values of the dissipation are computed numerically from the constitutive model. For simplification we set the co-ordinate x to $1_o/2$ and the initial length 1_o of the specimen to 10 mm. The simulations are carried out for different displacement amplitudes and mean values. The loading frequency $\omega/2\pi$ is varied between 10^{-3} Hz and 10^1 Hz.

In Fig. 10, $\vartheta_M(1_o/2)$ is plotted against the frequency of the loading process. Solid lines correspond to an amplitude of 0.2 mm, dotted lines to 0.5 mm and dashed lines to 1 mm. Different symbols represent different values of u_o. The simulations show that the temperature increase remains below 1 K if the applied loading frequencies remain below 2×10^{-1} Hz. Furthermore we see that the temperature increase depends on the displacement amplitude as well as on its mean value. If the frequencies are larger that 10 Hz, the temperature increase

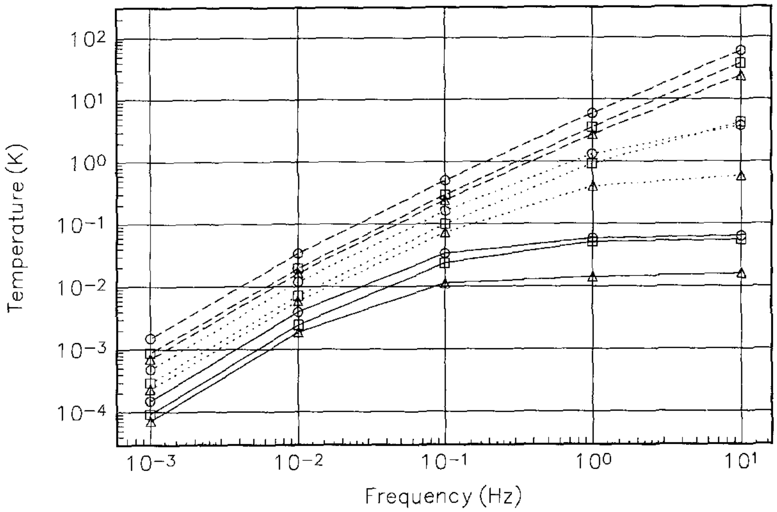

Fig. 10. Increase of the mean temperature as a function of the displacement frequency ($\Delta u = 0.2$ mm, 0.5 mm, 1 mm) ○ $\hat{=}\ u_o = -0.1$ mm, ■ $\hat{=}\ u_o = 0$, $\Delta\ \hat{=}\ u_o = 0.1$ mm

becomes larger than 10 K. In this case, the assumption of temperature independent material parameters no longer applies, and the theory has to be improved.

By this example we have demonstrated that the developed constitutive theory in combination with the fundamental laws of thermodynamics facilitates the representation of thermomechanical heating phenomena, which are caused by inelastic deformations. Clearly, if the proposed theory will be applied to compute thermomechanical heating phenomena in the high frequency range, the temperature dependence of the material properties should be investigated experimentally.

6 Conclusions

In this paper a three-dimensional internal variable theory of thermoviscoplasticity for elastomeric materials is proposed. The method to develop the theory starts with the formulation of a uniaxial rheological model which incorporates the experimentally observed phenomena and satisfies the dissipation inequality. The basic idea is an additive decomposition of the total stress into a rate independent equilibrium stress and a nonlinear rate dependent overstress. The equilibrium stress is decomposed into a sum of two terms as well. The first term is a nonlinear function of the total strain which is attributed to the elastic behaviour of the material, and the second term is a history dependent part which represents the equilibrium hysteresis effects. Motivated by the rheological model, the overstress and the hysteretic part of the equilibrium stress are determined by nonlinear elasticity relations which depend on elastic strains. In order to satisfy the dissipation principle, the evolution equations for the inelastic strains are formulated in consideration of the Clausius Duhem inequality.

After this, the physical ideas of the rheological model are successfully generalised to the three-dimensional geometric nonlinear case. To this end, two multiplicative decompositions of the deformation gradient into elastic and inelastic parts are introduced, and a set of physically reasonable stress- and strain measures is obtained in consideration of the concept of dual variables. The constitutive equations for the overstress and the hysteretic part of the equilibrium stress are formulated with respect to their associated intermediate configurations, and the elastic part of the equilibrium stress is formulated on the reference configuration. In order to simplify the representation of inelastic bulk and distortional effects, kinematic decompositions of the deformation gradient are also introduced.

A comparison between a large set of experimental data and numerical simulations illustrates that the three-dimensional theory reproduces the mechanical behaviour of carbon black reinforced rubber at room temperature very well. In particular, nonlinear elasticity in combination with nonlinear rate-dependence and equilibrium hysteresis effects are described. By an example it is demonstrated that the constitutive theory also represents dissipative heating phenomena which result from inelastic deformations.

In order to extend the constitutive theory, the temperature dependence of the material properties should be investigated experimentally. To this end, isothermal monotonic and cyclic experiments at different temperatures as well as suitable experiments to analyse the dissipation properties must be carried out. In addition, thermal expansion effects should be incorporated into the theory. Physically, this work is only a first step up to a complete representation of the complicated thermomechanical behaviour of elastomers.

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