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A pseudo-elastic model for loading, partial unloading and reloading of particle-reinforced rubber

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Abstract

Particle-reinforced rubbers exhibit a marked stress softening during unloading after loading in uniaxial tension tests, i.e. the stress on unloading is significantly less than that on loading at the same stretch. This hysteretic behaviour is not accounted for when the mechanical properties are represented in terms of a strain-energy function, i.e. if the material is modelled as hyperelastic. In this paper a theory of *pseudo-elasticity* is used to model loading, partial or complete unloading and the subsequent reloading and unloading of reinforced rubber. The basis of the model is the inclusion in the energy function of a variable that enables the energy function to be changed as the deformation path changes between loading, partial unloading, reloading and any further unloading. The dissipation of energy, i.e. the difference between the energy input during loading and the energy returned on unloading is accounted for in the model by the use of a *dissipation function*, the form of which changes between unloading, reloading and subsequent unloading.

Keywords: Pseudo-elasticity; Particle-reinforced rubber; Dissipation function; Hysteretic response

1. Introduction

During the last decade there has been a considerable growth in interest in modelling the mechanical response of rubbery polymers, and more particularly of particle-filled rubbers. This interest has been stimulated by numerous industrial applications of rubbers (vibration isolators, vehicle tyres, seals and shock absorbers, for example) and the availability of computational facilities suitable for running complex models in finite element software.

Many contributions have aimed to model inelastic behaviour of rubber such as the stress softening associated with the Mullins effect. These are mainly rate and time-independent models based on the use of damage theory. Representative examples of these works are the papers by Govindjee and Simo (1991, 1992a,b), extended to allow for viscoelasticity (1992b), Johnson and Beatty (1993, 1995), Lion (1996),

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Huntley et al. (1997), Kaliske and Rothert (1998), Ogden and Roxburgh (1999a,b), Beatty and Krishnaswamy (2000) and Dorfmann et al. (2002). There are also many contributions dealing with time and/or rate effects and stress–strain cycling involving hysteresis. Representative works from a lengthy list include Johnson et al. (1995), Drozdov (1996), Drozdov and Dorfmann (2001), Ha and Schapery (1998), Reese and Govindjee (1998), Bergström and Boyce (1998, 2000), Miehe and Keck (2000), Wu and Liechti (2000) and Yang et al. (2000). The collections of papers contained in the proceedings of the first two European conferences on constitutive models for rubber are also valuable sources of reference: Dorfmann and Muhr (1999) and Besdo et al. (2001). Both phenomenological and micro-structural models are represented in these contributions and for more detailed references we refer to the above-cited works.

The present paper focuses on the *quasi-static* modelling of the inelastic response of particle-reinforced rubber. In particular, we are concerned with the hysteretic cycles associated with partial unloading and reloading (at constant temperature) following loading after appropriate pre-conditioning aimed at eliminating the Mullins effect. Our starting point is the pseudo-elasticity theory of Ogden and Roxburgh (1999a), which was used to model the Mullins effect. It is adapted so as to model the hysteretic cycles mentioned above. While the theory is applicable to three-dimensional deformations, the details are described primarily for the simple tension specialization. Simple tension experiments on a 60 phr carbon black-filled rubber have been performed for loading, partial unloading, reloading and subsequent unloading in order to test the theory.

The paper is organized as follows. In Section 2 we summarize the required equations of non-linear elasticity, first for three dimensions and then for the appropriate homogeneous uniaxial specialization. In Section 3 the corresponding theory of pseudo-elasticity is outlined. In Section 4, first the specific model of Ogden and Roxburgh (1999a), with some modification, is reviewed and then adapted so as to capture the partial unloading–reloading–unloading response. Section 5.1 contains a brief discussion of the experimental results that are used as the basis for fitting the model. The elastic strain-energy function employed for describing the loading response (after pre-conditioning) is given in Section 5.2, and then the theory of Section 4 is used to fit the actual data.

2. Basic equations

For full details of the relevant theory of elasticity summarized in this section the reader is referred to, for example, Ogden (1984, 2001) and Holzapfel (2000).

We consider a rubberlike solid whose deformation is completely described by the deformation gradient tensor $\mathbf{F}$. The polar decompositions of the deformation gradient gives

$$\mathbf{F} = \mathbf{R}\mathbf{U} = \mathbf{V}\mathbf{R}, \tag{1}$$

where $\mathbf{R}$ is a proper orthogonal tensor and $\mathbf{U}, \mathbf{V}$ are positive definite and symmetric tensors (the right and left stretch tensors, respectively).

The *spectral decomposition* of the right stretch tensor $\mathbf{U}$ gives

$$\mathbf{U} = \sum_{i=1}^{3} \lambda_i \mathbf{u}^{(i)} \otimes \mathbf{u}^{(i)}, \tag{2}$$

where the principal stretches $\lambda_i > 0$, $i \in \{1, 2, 3\}$, are the eigenvalues of $\mathbf{U}$, $\mathbf{u}^{(i)}$ are the (unit) eigenvectors, and $\otimes$ denotes the tensor product. From the incompressibility condition $\det \mathbf{F} = 1$ and from (1) and (2) it follows that

$$\lambda_1 \lambda_2 \lambda_3 = 1. \tag{3}$$

2.1. Hyperelasticity

For hyperelastic solids there exists a strain-energy function, denoted $W = W(\mathbf{F})$. The associated nominal stress tensor for incompressible elastic material, denoted $\mathbf{S}$, is then given by

$$\mathbf{S} = \frac{\partial W}{\partial \mathbf{F}} - p\mathbf{F}^{-1}, \quad \det \mathbf{F} = 1, \tag{4}$$

where p is a Lagrange multiplier associated with the constraint (3) and represents an arbitrary hydrostatic pressure. The Cauchy stress tensor $\boldsymbol{\sigma}$ is given by

$$\boldsymbol{\sigma} = \mathbf{F}\mathbf{S} = \mathbf{F}\frac{\partial W}{\partial \mathbf{F}} - p\mathbf{I}, \quad \det \mathbf{F} = 1, \tag{5}$$

where $\mathbf{I}$ is the identity tensor.

The elastic stored energy is required to be objective. Therefore, for all rotations $\mathbf{Q}$ we have

$$W(\mathbf{QF}) = W(\mathbf{F}). \tag{6}$$

Using the polar decomposition (1) and the choice $\mathbf{Q} = \mathbf{R}^{\mathrm{T}}$ in (6) gives

$$W(\mathbf{F}) = W(\mathbf{U}). \tag{7}$$

Thus, W depends on $\mathbf{F}$ only through the stretch tensor $\mathbf{U}$. The (symmetric) Biot stress tensor $\mathbf{T}$ is then defined by

$$\mathbf{T} = \frac{\partial W}{\partial \mathbf{U}} - p\mathbf{U}^{-1}, \quad \det \mathbf{U} = 1. \tag{8}$$

2.1.1. Isotropic hyperelasticity

We now consider *isotropic* elastic materials, for which we have the restriction

$$W(\mathbf{FQ}) = W(\mathbf{F}) \tag{9}$$

for all rotations $\mathbf{Q}$. Bearing in mind that the $\mathbf{Q}$'s appearing in (6) and (9) are independent the combination of these two equations yields

$$W(\mathbf{QUQ}^{\mathrm{T}}) = W(\mathbf{U}) \tag{10}$$

for all rotations $\mathbf{Q}$, or, equivalently, $W(\mathbf{QVQ}^{\mathrm{T}}) = W(\mathbf{V})$. Eq. (10) states that W is an isotropic function of $\mathbf{U}$. It follows from the spectral decomposition (2) that W depends on $\mathbf{U}$ only through the principal stretches $\lambda_1, \lambda_2, \lambda_3$. To avoid introducing additional notation we express this dependence as $W(\lambda_1, \lambda_2, \lambda_3)$; by selecting appropriate values for $\mathbf{Q}$ in (10) we may deduce that W depends symmetrically on $\lambda_1, \lambda_2, \lambda_3$, i.e.

$$W(\lambda_1, \lambda_2, \lambda_3) = W(\lambda_1, \lambda_3, \lambda_2) = W(\lambda_2, \lambda_1, \lambda_3). \tag{11}$$

Consequences of isotropy are that $\mathbf{S} = \mathbf{TR}^{\mathrm{T}}$ and that $\mathbf{T}$ is coaxial with $\mathbf{U}$ and hence, in parallel with (2), we have

$$\mathbf{T} = \sum_{i=1}^{3} t_i \mathbf{u}^{(i)} \otimes \mathbf{u}^{(i)}, \tag{12}$$

where t_i, $i \in \{1, 2, 3\}$, are the principal Biot stresses, given by

$$t_i = \frac{\partial W}{\partial \lambda_i} - p\lambda_i^{-1}, \quad \lambda_1\lambda_2\lambda_3 = 1. \tag{13}$$

We also note the connection between the Cauchy stress and $\mathbf{T}$ in the form

$$\mathbf{R}^{\mathrm{T}}\boldsymbol{\sigma}\mathbf{R} = \mathbf{UT} = \mathbf{TU} = \sum_{i=1}^{3} \lambda_i t_i \mathbf{u}^{(i)} \otimes \mathbf{u}^{(i)}, \tag{14}$$

from which it follows that the principal Cauchy stresses σ_i, $i \in \{1,2,3\}$, are given by

$$\sigma_i = \lambda_i t_i = \lambda_i \frac{\partial W}{\partial \lambda_i} - p, \tag{15}$$

where p is the arbitrary hydrostatic pressure appearing in (4). There is no sum over i in (15).

2.1.2. Simple tension and compression

The isotropic strain-energy function W depends on the principal stretches according to (11). On use of the incompressibility constraint (3), it can be written in terms of two independent stretches. Thus, we write

$$\widehat{W}(\lambda_1, \lambda_2) = W(\lambda_1, \lambda_2, \lambda_1^{-1}\lambda_2^{-1}), \tag{16}$$

which is symmetric in λ_1 and λ_2. Then, from (15) we obtain the Cauchy stress differences

$$\sigma_1 - \sigma_3 = \lambda_1 \frac{\partial \widehat{W}}{\partial \lambda_1}, \quad \sigma_2 - \sigma_3 = \lambda_1 \frac{\partial \widehat{W}}{\partial \lambda_2}. \tag{17}$$

Eqs. (17) provide a basis for characterizing the form of the energy function using biaxial tests in which λ_1 and λ_2 are varied independently. For this purpose and without loss of generality we can set σ_3 equal to zero.

For the simple tension (or compression) specialization we take $\lambda_2 = \lambda_3$, and use the notation

$$\lambda_1 = \lambda, \quad \lambda_2 = \lambda^{-1/2}. \tag{18}$$

The strain energy then depends on the one remaining independent stretch, and we write

$$\widetilde{W}(\lambda) = \widehat{W}(\lambda, \lambda^{-1/2}). \tag{19}$$

Then, the Cauchy stress associated with λ_1 is

$$\sigma = \sigma_1 = \lambda \frac{\mathrm{d}\widetilde{W}(\lambda)}{\mathrm{d}\lambda} \tag{20}$$

and the corresponding nominal (or Biot) stress is

$$t = \frac{\sigma}{\lambda} = \frac{\mathrm{d}\widetilde{W}(\lambda)}{\mathrm{d}\lambda}. \tag{21}$$

3. Pseudo-elasticity

3.1. Basic equations

In the theory of pseudo-elasticity developed by Ogden and Roxburgh (1999a) the strain-energy function $W(\mathbf{F})$ appropriate for elasticity theory is modified by incorporating an additional variable η into the function. Thus, we write

$$W = W(\mathbf{F}, \eta). \tag{22}$$

In the context of the Mullins effect, which is related to material damage, η is referred to as a damage or softening variable. The inclusion of η provides a means of changing the form of the energy function during the deformation process and hence changing the character of the material properties. In general, the overall response of the material is then no longer elastic and $W(\mathbf{F},\eta)$ is referred to as a *pseudo-energy function*. The resulting theory is referred to as *pseudo-elasticity theory*. In this section we summarize the main ingredients of the theory.

The variable η may be active or inactive and a change from active to inactive (or conversely) effects a change in the material properties. This change may be induced, for example, when unloading is initiated.

If η is inactive we set it to the constant value unity and write

$$W_0(\mathbf{F}) = W(\mathbf{F},1) \tag{23}$$

for the resulting strain-energy function. For an incompressible material the associated nominal stress is denoted $\mathbf{S}_0$ and is given by

$$\mathbf{S}_0 = \frac{\partial W_0}{\partial \mathbf{F}}(\mathbf{F}) - p_0\mathbf{F}^{-1}, \quad \det \mathbf{F} = 1. \tag{24}$$

In (24) and in what follows the zero subscript is associated with the situation in which η is inactive.

If η is active we take it to depend on $\mathbf{F}$. The nominal stress is then given by

$$\mathbf{S} = \frac{\partial W}{\partial \mathbf{F}}(\mathbf{F},\eta) + \frac{\partial W}{\partial \eta}(\mathbf{F},\eta)\frac{\partial \eta}{\partial \mathbf{F}}(\mathbf{F}) - p\mathbf{F}^{-1}, \quad \det \mathbf{F} = 1. \tag{25}$$

Following Ogden and Roxburgh (1999a) we take η to be given implicitly by the constraint

$$\frac{\partial W}{\partial \eta}(\mathbf{F},\eta) = 0 \tag{26}$$

so that the nominal stress then has the form

$$\mathbf{S} = \frac{\partial W}{\partial \mathbf{F}}(\mathbf{F},\eta) - p\mathbf{F}^{-1}, \quad \det \mathbf{F} = 1 \tag{27}$$

whether or not η is active, where, when η is active the right-hand side is evaluated for η given by (26).

Under the constraint $\det \mathbf{F} = 1$, Eq. (26) defines a hypersurface in the resulting nine-dimensional $(\mathbf{F},\eta)$-space to which values of η must be restricted. The hypersurface (26) identifies stationary points of $W(\mathbf{F},\eta)$ with respect to η. If η is defined uniquely in terms of $\mathbf{F}$ we may write the solution formally as

$$\eta = \eta_e(\mathbf{F}), \tag{28}$$

and we then use the notation w for the resulting (unique) strain-energy function. Thus,

$$w(\mathbf{F}) \equiv W(\mathbf{F},\eta_e(\mathbf{F})). \tag{29}$$

Thus far we have not specified the form of the dependence of W on η, or, more particularly, the form of the function $\eta_e(\mathbf{F})$ in (28), i.e. we have not specified a particular model within the general framework of pseudo-elasticity. Appropriate specification will be made in Section 4.

3.2. Isotropic material response

When specialized to isotropic response (relative to the selected reference configuration) the pseudo-elastic energy function (22) takes the form

$$W(\lambda_1,\lambda_2,\lambda_3,\eta), \tag{30}$$

where $(\lambda_1,\lambda_2,\lambda_3)$ are the principal stretches associated with the deformation from the reference configuration. As in Section 2.1.1, W is a symmetric function of the stretches, which are subject to the incompressibility constraint (3).

The principal Cauchy stresses σ_i are given by

$$\sigma_i = \lambda_i \frac{\partial W}{\partial \lambda_i} - p, \quad i \in \{1, 2, 3\}, \tag{31}$$

as in (15), but (31) applies whether or not η is active. Eq. (26) specializes to

$$\frac{\partial W}{\partial \eta}(\lambda_1, \lambda_2, \lambda_3, \eta) = 0, \tag{32}$$

which gives η implicitly in terms of the stretches.

Since the material is incompressible it is convenient to define the modified pseudo-energy function $\widehat{W}(\lambda_1, \lambda_2, \eta)$ by

$$\widehat{W}(\lambda_1, \lambda_2, \eta) = W(\lambda_1, \lambda_2, \lambda_1^{-1}\lambda_2^{-1}, \eta), \tag{33}$$

extending the notation used in (16). Then, on elimination of p from (31),

$$\sigma_1 - \sigma_3 = \lambda_1 \widehat{W}_1, \quad \sigma_2 - \sigma_3 = \lambda_2 \widehat{W}_2, \tag{34}$$

where $\widehat{W}_1$ and $\widehat{W}_2$ denote the partial derivatives of $\widehat{W}$ with respect to λ_1 and λ_2, respectively. Eq. (32) is then modified to

$$\frac{\partial \widehat{W}}{\partial \eta}(\lambda_1, \lambda_2, \eta) = 0 \tag{35}$$

and hence η is now given implicitly in terms of λ_1 and λ_2 only.

We define the function $\widehat{W}_0(\lambda_1, \lambda_2)$ via

$$\widehat{W}_0(\lambda_1, \lambda_2) \equiv \widehat{W}(\lambda_1, \lambda_2, 1), \tag{36}$$

which is the isotropic specialization of (23). This is the energy function of an elastic material for which η is inactive. From (34) the specialization (36) yields the stresses

$$\sigma_{01} - \sigma_{03} = \lambda_1 \widehat{W}_{01}, \quad \sigma_{02} - \sigma_{03} = \lambda_2 \widehat{W}_{02}, \tag{37}$$

where the subscript zero again refers to a deformation path on which η is not active and (35) is not operative. A subscript 1 (respectively 2) following the subscript 0 on $\widehat{W}$ indicates partial differentiation with respect to λ_1 (respectively λ_2).

For compatibility with the classical theory $\widehat{W}_0(\lambda_1, \lambda_2)$ must satisfy

$$\widehat{W}_0(1, 1) = 0, \quad \widehat{W}_{0\alpha}(1, 1) = 0, \quad \widehat{W}_{012}(1, 1) = 2\mu, \quad \widehat{W}_{0\alpha\alpha}(1, 1) = 4\mu, \tag{38}$$

where μ (> 0) is the shear modulus of the material in the reference configuration and the index α takes the value 1 or 2.

When η is active we suppose that Eq. (35) can be solved explicitly for η and, using the notation from (28), we write

$$\eta = \eta_e(\lambda_1, \lambda_2) = \eta_e(\lambda_2, \lambda_1). \tag{39}$$

Then, an energy function for active η, symmetrical in (λ_1, λ_2) and denoted $\hat{w}(\lambda_1, \lambda_2)$, may be defined by

$$\hat{w}(\lambda_1, \lambda_2) \equiv \widehat{W}(\lambda_1, \lambda_2, \eta_e(\lambda_1, \lambda_2)). \tag{40}$$

From Eqs. (34), (35) and (40) it follows that

$$\sigma_\alpha - \sigma_3 = \lambda_\alpha \partial\hat{w}/\partial\lambda_\alpha = \lambda_\alpha \partial\widehat{W}/\partial\lambda_\alpha, \quad \alpha = 1, 2. \tag{41}$$

3.2.1. Simple tension

As in Section 2.1.2, for simple tension we take $\sigma_2 = \sigma_3 = 0$ and write $\sigma_1 = \sigma$. We also write $\lambda_1 = \lambda$, so that $\lambda_2 = \lambda_3 = \lambda^{-1/2}$, and define $\widetilde{W}$ by

$$\widetilde{W}(\lambda, \eta) \equiv \widehat{W}(\lambda, \lambda^{-1/2}, \eta). \tag{42}$$

Eqs. (41) and (35) then specialize to

$$\sigma = \lambda \widetilde{W}_\lambda(\lambda, \eta) \equiv \lambda t, \quad \widetilde{W}_\eta(\lambda, \eta) = 0, \tag{43}$$

wherein the principal Biot stress $t\ (= t_1)$ is defined and the subscripts signify partial derivatives.

By defining

$$\widetilde{W}_0(\lambda) = \widetilde{W}(\lambda, 1), \tag{44}$$

we may deduce from (38) the specializations

$$\widetilde{W}_0(1) = \widetilde{W}_0'(1) = 0, \quad \widetilde{W}_0''(1) = 3\mu, \tag{45}$$

where the prime signifies differentiation with respect to λ.

This simple tension specialization will be examined in detail in connection with the description of stress softening in Section 4.

4. A model for unloading and reloading

In this section we use a simple form for the pseudo-elastic constitutive law that was used previously by Ogden and Roxburgh (1999a) to model the idealized Mullins effect. Here, however, it is assumed that the Mullins effect is not present (having been removed by pre-conditioning) and we are concerned with modelling the hysteresis associated with loading–unloading cycles, and more particularly with partial unloading and reloading. The material is again taken to be incompressible and isotropic and we use a pseudo-energy function to represent unloading in the form

$$\widehat{W}(\lambda_1, \lambda_2, \eta) = \eta \widehat{W}_0(\lambda_1, \lambda_2) + \phi(\eta), \quad \phi(1) = 0, \tag{46}$$

based on (λ_1, λ_2)-space. From (37), (41) and (46), the Cauchy stress differences are calculated as

$$\sigma_\alpha - \sigma_3 = \eta \lambda_\alpha \widehat{W}_{0\alpha} = \eta(\sigma_{0\alpha} - \sigma_{03}), \quad \alpha = 1, 2, \tag{47}$$

and Eq. (35) becomes

$$\phi'(\eta) = -\widehat{W}_0(\lambda_1, \lambda_2), \tag{48}$$

which, implicitly, defines the parameter η in terms of the stretches.

We define a loading path in (λ_1, λ_2)-space as a path starting from $(1, 1)$ on which $\widehat{W}_0$ is increasing. As mentioned by Ogden and Roxburgh (1999a), for many standard forms of strain-energy function $\widehat{W}_0$ is increasing along any straight line path from $(1, 1)$ and contours of constant energy are actually convex in (λ_1, λ_2)-space.

4.1. Uniaxial unloading

On the basis of the equations in Section 3.2.1 the specializations of the above equations for simple tension are

$$\widetilde{W}(\lambda, \eta) = \eta \widetilde{W}_0(\lambda) + \phi(\eta), \quad \phi(1) = 0, \tag{49}$$

and, in terms of the Biot stress t,

$$t = \eta \widetilde{W}_0'(\lambda) = \eta t_0, \tag{50}$$

where t_0 is the Biot stress on the loading path at the same value of λ. For (50) to predict stress softening on unloading, at the start of which η is switched on, it is clear that we must have $\eta \leqslant 1$ on the unloading path, with equality only at the point where unloading begins. Here, as in Ogden and Roxburgh (1999a), we take $\eta > 0$, so that t remains positive on unloading until $\lambda = 1$ is reached. The occurrence of residual strains is therefore excluded in the present treatment.

The simple tension specialization of (48) is

$$\phi'(\eta) = -\widetilde{W}_0(\lambda). \tag{51}$$

On differentiation of (51) with respect to λ we obtain

$$\phi''(\eta)\frac{\mathrm{d}\eta}{\mathrm{d}\lambda} = -\widetilde{W}_0'(\lambda). \tag{52}$$

In view of the stress softening requirement discussed above we associate unloading with decreasing η. Since $t_0 \equiv \widetilde{W}_0'(\lambda) > 0$ for $\lambda > 1$ it follows from (52) that

$$\phi''(\eta) < 0, \tag{53}$$

and we assume henceforth that this inequality holds. We deduce that $\phi'(\eta)$ is a monotonic decreasing function of η and hence that η is uniquely determined from (51) as a function of $\widetilde{W}_0(\lambda)$.

It is important to point out that the value of η derived from (51) depends on the value of the principal stretch, λ_m say, attained on the loading path, as well as on the specific forms of $\widetilde{W}_0(\lambda)$ and $\phi(\eta)$ employed. Since $\eta = 1$ at any point on the loading path from which unloading is initiated, it follows from Eqs. (49) and (51) that

$$\phi'(1) = -\widetilde{W}_0(\lambda_m) \equiv -W_m, \tag{54}$$

wherein the notation W_m is defined. This is the current maximum value of the energy achieved on the loading path. In accordance with the properties of $\widetilde{W}_0$, W_m increases along a loading path. In view of (54), the function ϕ depends (implicitly) on the point from which unloading begins through the energy expended on the loading path up to that point.

When the material is fully unloaded, with $\lambda = 1$, η attains its minimum value, $\eta_{\min}$ say. This is determined by inserting these values into Eq. (51) to give, using the first equation in (45),

$$\phi'(\eta_{\min}) = -\widetilde{W}_0(1) = 0. \tag{55}$$

Since the function ϕ depends on the point where unloading begins then so does $\eta_{\min}$, that is it depends, though W_m, on the value of λ_m. The pseudo-energy function (49) has the residual value

$$\widetilde{W}(1, \eta_{\min}) = \phi(\eta_{\min}). \tag{56}$$

Thus, the residual (non-recoverable) energy $\phi(\eta_{\min})$ may be interpreted as a measure of the energy dissipated in the material during the loading–unloading cycle. In simple tension $\phi(\eta_{\min})$ is the area between the primary loading curve and the relevant unloading curve. It is therefore appropriate to refer to ϕ as a *dissipation function*.

Unloading may take place from any point on the loading path, and the start of unloading is taken as the signal for η to be activated, as mentioned above.

In order to satisfy the above requirements, we select the dissipation function ϕ to have the form

$$-\phi'(\eta) = m \tanh^{-1}[r(\eta - 1)] + W_m, \tag{57}$$

where r and m/μ are dimensionless positive material parameters, μ being the shear modulus appearing in (45). Note that this form of ϕ' differs from that used by Ogden and Roxburgh (1999a). From Eq. (57) we arrive, after a few minor manipulations, at

$$1 - \eta = \frac{1}{r} \tanh\left[\frac{W_m - \widetilde{W}_0(\lambda)}{m}\right]. \tag{58}$$

The minimum value $\eta_{\min}$ of the variable η is given for $\lambda = 1$, i.e. in the unstressed configuration, by

$$\eta_{\min} = 1 - \frac{1}{r} \tanh\left[\frac{W_m}{m}\right]. \tag{59}$$

Finally, integration of Eq. (57) gives ϕ explicitly in terms of the variable η in the form

$$\phi(\eta) = -m(\eta - 1)\tanh^{-1}[r(\eta - 1)] - W_m(\eta - 1) - \frac{m}{2r}\log[1 - r^2(\eta - 1)^2]. \tag{60}$$

4.2. Uniaxial reloading

During unloading the value of η is a monotonic function decreasing from its initial value 1 to its minimum value $\eta_{\min}$. Now, suppose that at a specific value of λ, λ_1 say, load reversal occurs, i.e. the material is again subjected to loading. The corresponding value of η is η_1, say, which is kept constant during the reloading phase. The pseudo-energy function for reloading is taken to have the form

$$\widetilde{W}_{\mathrm{r}}(\lambda, \eta_{\mathrm{r}}) = \eta_{\mathrm{r}} \widetilde{W}_0(\lambda) + \phi_{\mathrm{r}}(\eta_{\mathrm{r}}), \tag{61}$$

where, for consistency, the variable η_{r} must increase from η_1 to the final value 1 during reloading and

$$\phi_{\mathrm{r}}(\eta_1) = \phi(\eta_1). \tag{62}$$

Once this final value $\eta_{\mathrm{r}} = 1$ is reached the material response switches from the unloading to the loading path. The subscript 'r' is used to emphasize that (61) applies only during reloading.

To satisfy Eq. (26) we must have

$$\phi'_{\mathrm{r}}(\eta_{\mathrm{r}}) = -\widetilde{W}_0(\lambda), \tag{63}$$

which, when evaluated at the start of reloading, gives

$$\phi'_{\mathrm{r}}(\eta_1) = -\widetilde{W}_0(\lambda_1). \tag{64}$$

A suitable expression of a monotonic increasing function to be used for η_{r}, having the same structure as (58), is

$$\frac{\eta_{\mathrm{r}}}{\eta_1} - 1 = \frac{(1 - \eta_1)}{\eta_1} \tanh\left[\frac{\widetilde{W}_0(\lambda) - \widetilde{W}_0(\lambda_1)}{a_1}\right], \tag{65}$$

where $\widetilde{W}_0(\lambda_1)$ is the total elastic energy stored in the material at the instant of load reversal and $a_1 = a(\widetilde{W}_0(\lambda_1))$ is a material parameter that reflects the changing material properties as reloading takes place from the (partially) unloaded configuration back to the loading curve. It is based on a function $a(\widetilde{W}_0(\lambda_1))$ of the energy $\widetilde{W}_0(\lambda_1)$. If the hysteretic response is interpreted in terms of recoverable damage then a_1 describes the recovery process. A short calculation enables the derivative of the dissipation function ϕ_{r} during reloading to be given as

$$\phi_r'(\eta_r) = -\widetilde{W}_0(\lambda) = -\widetilde{W}_0(\lambda_1) - a_1 \tanh^{-1}\left[\frac{\eta_r - \eta_1}{1 - \eta_1}\right]. \tag{66}$$

Note that $\phi_r''(\eta_r) < 0$. Integration of Eq. (66) provides an explicit form for $\phi_r(\eta_r)$, namely

$$\phi_r(\eta_r) = \phi(\eta_1) - (\eta_r - \eta_1)\widetilde{W}(\lambda_1) - a_1(\eta_r - \eta_1)\tanh^{-1}\left[\frac{\eta_r - \eta_1}{1 - \eta_1}\right] - \frac{1}{2}a_1(1 - \eta_1)\log\left[1 - \frac{(\eta_r - \eta_1)^2}{(1 - \eta_1)^2}\right]. \tag{67}$$

At $\lambda = \lambda_1$, Eqs. (60) and (67) give the same value and hence continuity of the pseudo-energy function is guaranteed. Eq. (61) with (67) gives the total energy per unit reference volume, i.e. the stored elastic energy together with the energy dissipated. If during reloading the value of η_r becomes 1 then the primary loading path is rejoined. However, if this value is not reached, a second unloading algorithm must be formulated and applied.

4.3. Second uniaxial unloading

Material unloading after reloading can be initiated from two different locations. The first one, described in Section 4.1, assumes that the material response lies on the initial loading path corresponding to $\eta = 1$. The second possibility, which is described here, arises when reloading is terminated before the primary loading path is reached, at a value $\eta_{ru} < 1$ of η_r. Let λ_{ru} be the value of λ at this point.

We assume that during this secondary unloading process the energy function is given by

$$\widetilde{W}_u(\lambda, \eta_u) = \eta_u \widetilde{W}_0(\lambda) + \phi_u(\eta_u), \tag{68}$$

where the subscript 'u' is associated with unloading following reloading. The energy dissipation is accounted for by the function $\phi_u(\eta_u)$, which, for continuity, satisfies

$$\phi_u(\eta_{ru}) = \phi_r(\eta_{ru}). \tag{69}$$

To satisfy Eq. (51) for the pseudo-energy function (68) we require

$$\phi_u'(\eta_u) = -\widetilde{W}_0(\lambda), \tag{70}$$

and at the start of unloading this becomes

$$\phi_u'(\eta_{ru}) = -\widetilde{W}_0(\lambda_{ru}). \tag{71}$$

Stress and energy continuity at the transition point requires that the initial value of η_u must be η_{ru}. We select the variable η_u so that

$$1 - \frac{\eta_u}{\eta_{ru}} = \tanh\left[\frac{\widetilde{W}_0(\lambda_{ru}) - \widetilde{W}_0(\lambda)}{b_{ru}}\right], \tag{72}$$

where $b_{ru} = b(\widetilde{W}_0(\lambda_{ru}))$ is a material parameter describing the hysteretic effect during the reloading and secondary unloading cycle. It is based on a function $b(\widetilde{W}_0(\lambda_{ru}))$ of the loading energy. Again, the structure of (72) is similar to (58).

The expression of the first derivative of the dissipation function representing secondary unloading is

$$\phi_u'(\eta_u) = -\widetilde{W}_0(\lambda) = b_{ru}\tanh^{-1}\left[\frac{\eta_{ru} - \eta_u}{\eta_{ru}}\right] - \widetilde{W}_0(\lambda_{ru}). \tag{73}$$

Then, after integration, the damage function to be used in expression (68) is obtained in the form

$$\phi_{\mathrm{u}}(\eta_{\mathrm{u}}) = \phi_{\mathrm{r}}(\eta_{\mathrm{ru}}) + (\eta_{\mathrm{ru}} - \eta_{\mathrm{u}})\widetilde{W}_0(\lambda_{\mathrm{ru}}) - b_{\mathrm{ru}}(\eta_{\mathrm{ru}} - \eta_{\mathrm{u}})\tanh^{-1}\left[\frac{\eta_{\mathrm{ru}} - \eta_{\mathrm{u}}}{\eta_{ru}}\right] - \frac{1}{2}b_{\mathrm{ru}}\eta_{\mathrm{ru}}\log\left[1 - \frac{(\eta_{ru} - \eta_{\mathrm{u}})^2}{\eta_{\mathrm{ru}}^2}\right]. \tag{74}$$

5. Numerical results

5.1. Experimental data

To assess the inelastic effect during the loading, partial unloading and reloading response of particle-reinforced elastomers, a series of uniaxial extension tests were carried out at a constant temperature. Dumbbell specimens were provided by SEMPERIT (Austria) and were used as received. The compound contains 60 phr of carbon black and is treated as a filled rubber.

The loading, partial unloading and reloading tests were performed at room temperature using a testing machine designed at the Institute of Physics (Vienna, Austria). A specimen was first subjected to six cycles of pre-conditioning up to a pre-selected extension of $\lambda = 3$. Pre-conditioning was performed in order to eliminate the influence of the Mullins effect and the results are shown in Fig. 1. To measure the longitudinal strain, two reflection lines (separated by a distance of 4 mm) were drawn in the central part of each specimen before loading. Changes in the distance between these lines were measured using a video-extensometer (which ensured the accuracy of about 1%). The tensile force was measured by using a standard loading cell and the nominal stress was determined as the ratio of the axial force to the cross-sectional area of a specimen (2 mm $\times$ 4 mm) in the stress-free state.

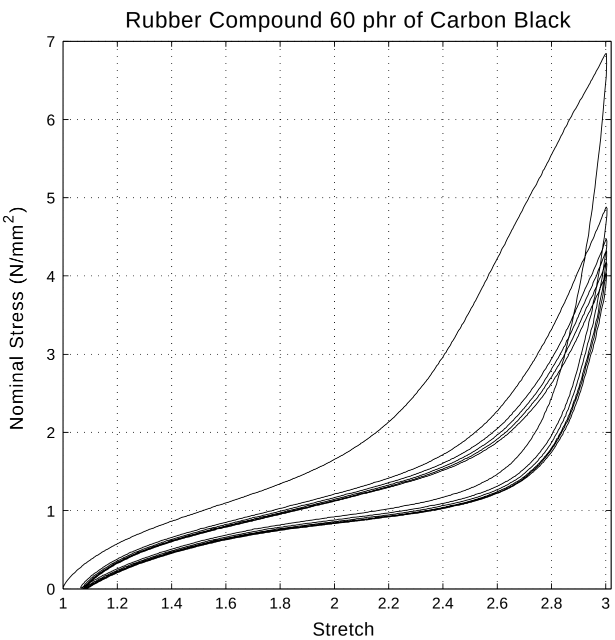

Fig. 1. Pre-conditioning of a particle-reinforced dumbbell specimen with 60 phr of carbon black.

After the initial six pre-conditioning loading–unloading cycles the stress strain response is essentially repeatable and no additional residual strain is generated beyond that produced in the first cycle. After the pre-conditioning cycles were completed the dumbbell specimen was stretched to $\lambda = 3$ which was taken as the starting point for evaluating the response of this compound to partial unloading and reloading. During the unloading phase, reloading is initiated at four different locations (see Fig. 2), corresponding to stretches of $\lambda = 2.5$, $\lambda = 2.0$, $\lambda = 1.5$ and $\lambda = 1.0$. It is interesting to note that, independently of the number of cycles to which the material is subjected, the reloading curve from $\lambda = 1$ follows the post–pre-conditioning loading curve. In Fig. 2 the residual strain accumulated during pre-conditioning has been subtracted in order for the graphs to be initiated at the origin. Therefore, the stretches where load reversal occurs are somewhat shifted to the left.

5.2. Material models

For the numerical results shown in this section the elastic strain energy suggested by Ogden (1972) has been used

$$W(\lambda_1, \lambda_2, \lambda_3) = \sum_{m=1}^{N} \mu_m(\lambda_1^{\alpha_m} + \lambda_2^{\alpha_m} + \lambda_3^{\alpha_m} - 3)/\alpha_m, \tag{75}$$

where α_m and μ_m are material constants to be determined by experiment and N is a positive integer. Most commonly N equals 3. For the simple tension and compression specialization equations (18) apply and the strain-energy function $\overline{W}_0(\lambda)$ is given by

$$\widetilde{W}_0(\lambda) = \sum_{m=1}^{N} \mu_m(\lambda^{\alpha_m} + 2\lambda^{-\alpha_m/2} - 3)/\alpha_m. \tag{76}$$

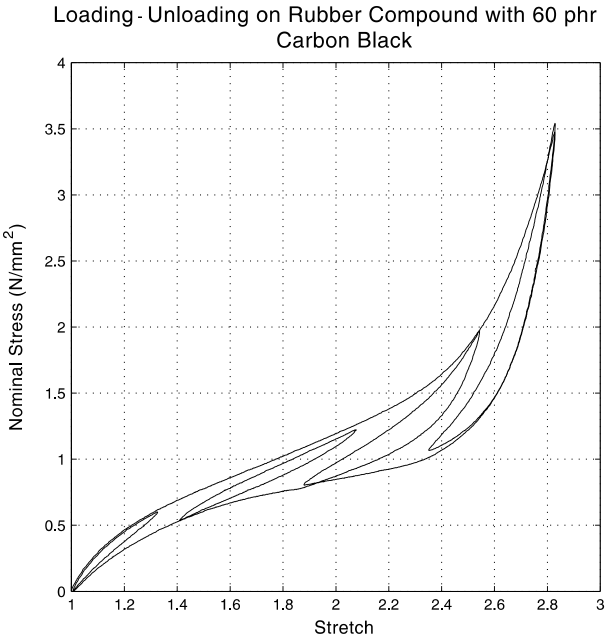

Fig. 2. Experimental data for partial reloading–unloading from the unloading path in simple tension of a rubber compound with 60 phr carbon black: nominal stress plotted against stretch.

A subscript 0 has been attached to $\widetilde{W}$ since we now use $\widetilde{W}_0(\lambda)$ to describe the loading path in simple tension. It follows that

$$t = t_1 = \sum_{m=1}^{N} \mu_m(\lambda^{\alpha_m} - \lambda^{-1/2\alpha_m}). \tag{77}$$

The constants must satisfy the requirement

$$\sum_{m=1}^{N} \mu_m\alpha_m = 2\mu, \tag{78}$$

where μ (> 0) is the shear modulus of the material in the natural configuration.

The non-linear iterative method known as the Levenberg–Marquardt algorithm (see, for example, Twizell and Ogden, 1983) is used for calculating the μ_i and α_i $(i = 1, 2, 3)$ in order to obtain a best fit of the primary loading curve shown in Fig. 2. These values are summarized in Table 1 and characterize the elastic strain energy in Eq. (75). Numerical results for loading, partial unloading and reloading are shown in Fig. 3. After initial loading up to $\lambda = 3$, unloading is initiated and the algorithm uses the formulations developed

Table 1
Summary of model parameters for loading curve of the 60 phr compound

Material model parameter, Ogden $N = 3$					
μ_1	α_1	μ_2	α_2	μ_3	α_3
−0.1680E−4	−12.0428	−0.5146	−4.4038	0.1722E−5	14.2651

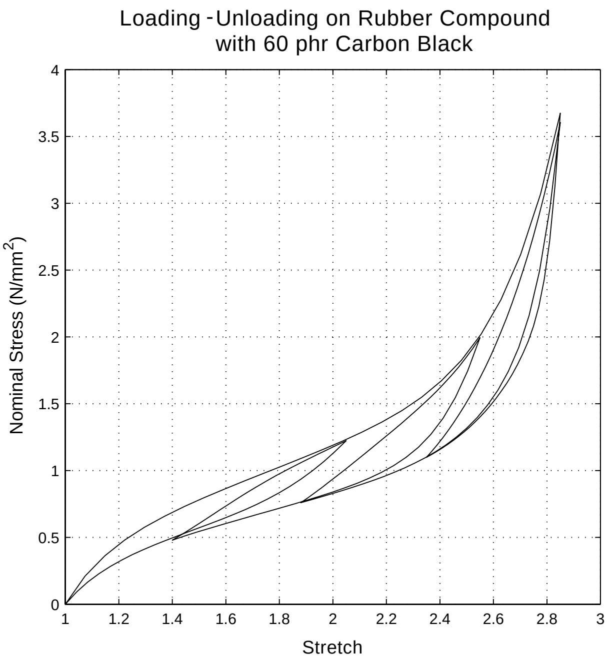

Fig. 3. Theoretical prediction of simple tension partial reloading–unloading behaviour using a pseudo-energy function: nominal stress plotted against stretch.

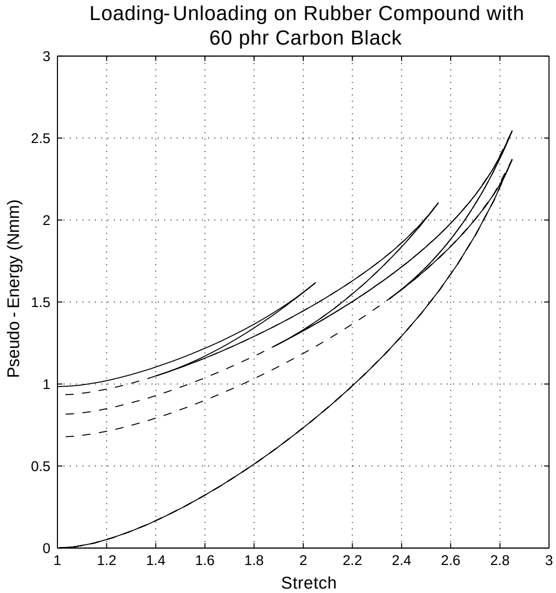

Fig. 4. Plot of the pseudo-energy function against the stretch during loading, unloading, reloading and unloading (continuous curves), showing continuity at the transition between (re)loading and unloading. The dashed curves correspond to complete unloading after each (re)loading. Their intercepts with the vertical axis quantify the energy dissipated, which increases with each reloading.

in Section 4.1. The parameters r and m in Eq. (58) are given as 3.3 (dimensionless) and 0.3 N mm, respectively. The pseudo-energy during unloading, given by Eq. (49), is fully determined using Eqs. (58), (76) and (60) and is depicted in Fig. 4. This figure shows the increase in energy during initial loading up to the instant of load reversal and it illustrates that during unloading the energy returned is less than that expended during loading, which is a consequence of the dissipation of energy due to hysteresis.

At the end of the first (partial) unloading path, at a stretch of about $\lambda = 2.4$, the material is reloaded up to the maximum extension. This reloading is modelled using the equations given in Section 4.2. Note from Fig. 3 that the initial loading curve has not been reached and the subsequent unloading curve is modelled on the basis of the equations in Section 4.3. A similar discussion applies for the other two reloading–unloading cycles shown in Fig. 3. In this representative example it was found sufficient to take $b_{\text{ru}} = b(\widetilde{W}_0(\lambda_{\text{ru}}))$ to be constant ($= 0.4$ N mm), i.e. the same value for each of the three unloading curves, while $a(\widetilde{W}_0(\lambda_1))$ is represented simply by $a(\widetilde{W}_0(\lambda_1)) = c_0 + c_1 \widetilde{W}_0(\lambda_1)$, where $c_0 = 0.2227$ N mm, $c_1 = 0.3723$, λ_1 being the value of λ at which reloading begins (different for each reloading path).

Continuity of the pseudo-energy at the transition points between unloading and reloading can be seen in Fig. 4. This figure also illustrates the extent of energy dissipation during cyclic loading and unloading. On each reloading–unloading cycle after partial unloading energy is dissipated and the energy returned on complete unloading is less than the energy expended during loading/reloading. The energy dissipated increases with each reloading–unloading cycle.

6. Discussion and conclusions

In this paper the theory of pseudo-elasticity, originally developed by Ogden and Roxburgh (1999a) to account for the Mullins effect, has been modified to develop a constitutive model for quasi-static loading, partial unloading, reloading and subsequent unloading of a rubber material with hysteretic response. The theory uses a deformation-dependent scalar parameter to modify the elastic strain-energy function to account for the change in material properties under large strains. A number of material parameters are included in the model to enable the fitting of simple tension data obtained. The dissipative character of the material is accounted for within the energy function by an additive dissipation term.

The suitability of the theory has been demonstrated for a particle-reinforced rubber (filled with 60 phr of carbon black) in the simple case of uniaxial cyclic loading and unloading in tension. However, the formulation presented here is also applicable to general (three-dimensional) deformations. The model does not allow for permanent set, the effect of which will be incorporated into the model in a separate paper.

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