Theory of thermohyperelasticity for near-incompressible elastomers

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Summary. Elastomers are often used in hot and confining environments in which thermomechanical properties are important. It appears that published constitutive models for elastomers are mostly limited to isothermal conditions. In this study, a *thermohyperelastic* constitutive model for near-incompressible elastomers is formulated in terms of the Helmholtz free energy density ϕ . Shear and volume aspects of the deformation are decoupled. Thermomechanical coupling occurs mostly as thermal expansion. Criteria for thermodynamic stability are derived in compact form. As illustration, a particular expression for ϕ is presented which represents the thermomechanical counterpart of the conventional two-term incompressible Mooney-Rivlin model. It is used to analyze several adiabatic problems in a rubber rod.

List of symbols

| A_i | matrices appearing in D |
|------------------------------------|--|
| c_e, \hat{c}_e, c_e' | specific heat at constant strain |
| С | Cauchy strain tensor |
| C_R | reduced Cauchy strain tensor |
| C_1, C_2 | coefficients of Mooney-Rivlin model |
| с | vectorial counterpart of C: VEC (C) |
| <i>c</i> ₂ | vectorial counterpart of C^2 : VEC (C^2) |
| D | isothermal tangent stiffness matrix |
| е | vectorial counterpart of ε : VEC (ε) |
| ē | deviatoric Lagrangian strain tensor |
| e _R | reduced deviatoric Lagrangian strain tensor |
| e | volume strain |
| e _T | reduced volume strain |
| f | thermal expansion function, $= [1 + \alpha (T - T_0)/3]^{-1}$ |
| $\boldsymbol{F}, \boldsymbol{F}_T$ | deformation gradient tensor |
| F_R | reduced deformation gradient tensor |
| H | Hessian matrix for the Gibbs free energy density ψ |
| <i>H</i> ′ | related to H |
| I_1, I_2, I_3 | invariants of C |
| I_{1R}, I_{2R}, I_{3R} | invariants of C_R |
| I, I9 | identity matrix |
| i | vectorial counterpart of I: VEC(I) |
| <i>J</i> , | determinant of F |
| J_R | determinant of F_R |
| J_T | determinant of F_T |
| J_{1}, J_{2} | invariants of $\varepsilon/J^{2/3}$ |
| J_{1R}, J_{2R} | invariants of $\varepsilon_R/J_R^{2/3}$ |
| k | thermal conductivity coefficient |
| K_1, K_2, K_3 | invariants of $\bar{e}/J^{2/3}$ |

| K_{1R}, K_{2R}, K_{3R} | invariants of $e_R/J^{2/3}$ |
|--|---|
| р | hydrostatic pressure |
| \$ | vectorial counterpart of stress σ : VEC (σ) |
| S | isotropic stress |
| Т | (absolute) temperature |
| T_0 | reference temperature |
| w | conventional (isothermal) strain energy density (per unit volume) |
| α | volumetric thermal expansion coefficient |
| β | thermal expansion vector |
| 8 | strain, Lagrangian strain |
| n | entropy density |
| х | isothermal bulk modulus |
| λ | Lagrange multiplier |
| λ_i | extension ratio |
| μ | shear modulus |
| χ | stability coefficient |
| Q | mass density |
| σ | stress, 2nd Piola-Kirchhoff stress |
| σ_i | principal stress |
| τ | Cauchy stress |
| τ _d | deviatoric Cauchy stress |
| $\phi, \phi_M, \phi_C, \phi_T, \phi_0$ | Helmholtz free energy density |
| ϕ_i | $\partial \phi / \partial I_i$ |
| ϕ_{ij} | $\partial^2 \phi / \partial I_i \partial I_j$ |
| ψ | Gibbs free energy density |
| δ(.) | variational operator |
| VEC (.) | vectorization operator |
| \otimes | operator for Kronecker product |

1 Introduction

1.1 General

Elastomers are often used in hot and confining environments, for example seals in automobile engines. Thermoelastic properties, described by *thermohyperelastic* constitutive models, may be important in such applications. In particular, thermal expansion may give rise to high stresses in areas of confinement. However, such behavior appears to have received limited study. For example, hyperelastic elements in finite element codes typically are suited only for isothermal conditions. The major thermohyperelastic models known to the authors are Dillon [1] and Shapery [2]. Both models involve Helmholtz energy density functions which in essence are series expansions in temperature and strain, and in any event do not emphasize near-incompressibility or associate thermal effects primarily with volume changes. Also of interest is the strain energy density function in the finite element code ABAQUS [3], which also accommodates thermal expansion.

Here we formulate a thermohyperelastic constitutive model for rubber, considered a nearincompressible elastomer. It is based on the assumption that temperature dependence is linear and its mechanical effect consists purely in thermal expansion. The resulting Helmholtz free energy density ϕ directly generalizes classical thermoelasticity. Criteria for thermodynamic stability are derived in compact form. Conditions for thermodynamic stability are presented. For illustration, a thermohyperelastic counterpart of the conventional two-term incompressible Mooney-Rivlin model is presented, and is used to analyze several adiabatic problems in a rubber rod.

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1.2 Summary of thermohyperelastic model

The Helmholtz free energy density (per unit mass) introduced in this investigation is as follows:

$$\phi(\varepsilon, T) = \phi_M(J_1, J_2) + \frac{\varkappa}{2\varrho} \left(f^3(T) J - 1 \right)^2 + c_e T \left[1 - \ln^4 \left(\frac{T}{T_0} \right) \right] + \phi_0.$$
(1)

The 2nd Piola-Kirchhoff stress and the entropy density are given by:

$$\sigma = \varrho \,\frac{\partial \phi}{\partial \varepsilon} = \,\varrho \,\frac{\partial \phi_1}{\partial \varepsilon} + \varkappa \big(f^3(T) \,J - 1\big) \,f^3(T) \,J C^{-1} \tag{2}$$

$$\eta = -\frac{\partial \phi}{\partial T} = \frac{\alpha \kappa}{\varrho} \left(f^3(T) J - 1 \right) J f^4(T) + c_e \ln \left(\frac{T}{T_0} \right)$$
(3)

where

$$f(T) = \frac{1}{1 + \frac{\alpha}{3} (T - T_0)}.$$
(4)

(Without loss of generality it has been tacitly assumed that $\eta = 0$ when $T = T_0$ and $\varepsilon = 0$.) Finally, the (hydrostatic) pressure is given by

$$p = -\frac{1}{3}\operatorname{trace}(\tau) = -\varkappa (f^{3}(T)J - 1)f^{3}(T).$$
(5)

This model retains many of the features of classical (linear, isotropic, small strain) thermoelasticity, to which it reduces in the small strain limit. Classical thermoelasticity is summarized in Appendix A. The primary nonlinearities are associated with the purely mechanical ("shear") aspects of the behavior, while thermal and thermomechanical effects are assumed to be linear. The material parameters α , \varkappa , c_{ρ} and ρ are considered to be known material constants unaffected by temperature. Purely thermal effects are represented in a linear manner by c_{e} (The thermal conductivity coefficient k can be included if conduction heat transfer is considered.) Coupling of the thermal and mechanical fields occurs through simple volumetric thermal expansion represented by a. Temperature rise induces strain, but not stress, in an unconstrained body. Finally, as in classical thermoelasticity, ϕ consists of four terms: a term ϕ_T depending purely on T; a "shear" term ϕ_M independent of temperature and volume change (T and J); a mixed term ϕ_c coupling temperature and volume through thermal expansion; and a constant term ϕ_0 corresponding to the reference configuration. The model is suited for near-incompressible materials involving small volume changes and a linear pressure-volume relation through \varkappa : in fact it is obtained by retaining the lowest order terms in a Taylor series expansion about J - 1 and $T = T_0$. The model affords a "smooth" transition from compressible to strictly incompressible behavior. Finally, for the sake of illustration, we introduce a particular form of ϕ representing the thermohyperelastic counterpart of the conventional two-term incompressible Mooney-Rivlin model:

$$\phi(\varepsilon, T) = C_1(J_1 - 3) + C_2(J_2 - 3) + \frac{\varkappa}{2\varrho} \left[f^3(T) J - 1 \right]^2 + c_e T \left[1 - \ln\left(\frac{T}{T_0}\right) \right] + \phi_0.$$
(6)

Criteria for thermodynamic stability of materials described by this model are derived in compact form. Finally, using values of C_1 , C_2 , α , \varkappa , c_e , and ϱ thought to be typical of natural rubber, graphical results are presented for several problems involving adiabatic response of a rubber rod.

2 Thermohyperelastic constitutive model

2.1 Isothermal conditions: compressible elastomers

Now let ε denote the Lagrangian strain tensor, and σ the 2nd Piola-Kirchhoff stress tensor. We also introduce the Cauchy strain tensor $C = 2\varepsilon + I$, with invariants I_1 , I_2 and $I_3 = J^2$. Here J is the determinant of the deformation gradient tensor F and represents volume change. Under isothermal conditions and assuming isotropy, $\varrho\phi$ may be equated with the strain energy function $w(I_1, I_2, J)$, which is traditionally used in the rubber literature (e.g., Nicholson and Nelson [4]). A well-known example is the two-term incompressible Mooney-Rivlin elastomer:

$$w = C_1(I_1 - 3) + C_2(I_2 - 3), \quad J = 1.$$
⁽⁷⁾

More generally, we assume that w = 0 if $\varepsilon = 0$.

We now invoke a formulation reviewed by Ogden [5]. Namely, if w is expressed in terms of $J_1 = I_1/J^{2/3}$, $J_2 = I_2/J^{4/3}$ and J, then

$$p = -\frac{\operatorname{trace}\left(\tau\right)}{3} = -\frac{\partial w}{\partial J} \tag{8}$$

in which p is the "true" hydrostatic pressure, and τ is the Cauchy stress given by

$$\tau = \frac{F\sigma F^T}{J}.\tag{9}$$

The bulk modulus \varkappa is defined by

$$\varkappa = -\frac{\partial p}{\partial J}.$$
(10)

For near-incompressible elastomers, it is assumed that J-1 is small, for which reason we consider a Taylor series through second order in J-1:

$$w = w_1 \left(\frac{\bar{e}}{J^{2/3}}\right) + a(J-1) + b(J-1)^2$$
(11)

in which \bar{e} is the deviator of the Lagrangian strain tensor, and a and b are as-yet-unknown constants. It is easily seen that, with a suitable choice of w_1 , the small strain case (cf. Appendix A with $T = T_0$) is recovered.

Now, from Eqs. (8), (11),

$$p = a + 2b(J - 1).$$
 (12)

But J = 1 implies that p = 0, and hence a = 0. Further, $b = \varkappa/2$ and hence the bulk modulus is a constant. Thus, to lowest order in J - 1,

$$w = w_1 + \frac{\varkappa}{2} (J-1)^2 \quad p = -\varkappa (J-1).$$
(13)

For later purposes we note that w may be rewritten as follows:

$$w = w_1 - p(J-1) - \frac{p^2}{2\varkappa}.$$
(14)

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The invariants of $\bar{e}/J^{2/3}$ are in fact:

$$K_1 = 0$$
 $K_2 = J_2 - \frac{J_1^2}{3}$ $K_3 = 1 - \frac{J_1 J_2}{3} + \frac{2J_1^3}{27}$ (15)

These relations can be inverted to furnish J_1 and J_2 in terms of K_2 and K_3 . It follows that any function of J_1 and J_2 can be viewed as a function of $\bar{e}/J^{2/3}$ and hence Eq. (13) can be expressed as $w = w_1(K_2, K_3) + \varkappa (J-1)^2/2$.

2.2 Isothermal conditions: incompressible and near incompressible elastomers

Incompressibility implies the constraint J = 1. For computational methods based on variational principles, such as the finite element method, w may be augmented as follows:

$$\hat{w} = w(J_1, J_2) - \lambda(J - 1) \tag{16}$$

in which λ is a Lagrange multiplier to be selected to render the total potential energy stationary under the incompressibility constraint (Oden [6]). Assuming independent variations of ε and λ , conditions for stationarity are

$$\sigma = \frac{\partial w}{\partial \varepsilon} - \lambda J C^{-1} \quad 0 = J - 1.$$
(17)

Now Eq. (8) implies that

$$p = \lambda. \tag{18}$$

Also, for near-incompressible elastomers, Eq. (14) is rewritten as

$$w = w_1(K_2, K_3) - p(J-1) - \frac{p^2}{2\varkappa}.$$
(19)

Now p and \bar{e} are regarded as variationally independent. The appropriate variational principle (cf. Cescotto et al. [7]) has a stationary rather than a minimum character. Conditions for stationarity are

$$\sigma = \frac{\partial w}{\partial \varepsilon} - pJC^{-1} \quad p = -\varkappa(J-1).$$
⁽²⁰⁾

We regard $s_1 = \partial w_1 / \partial \varepsilon$ as the generalized shear stress. It depends on the deviator (generalized shear component) of the Lagrangian strain. Further, recalling the transformation (9), it corresponds to the deviatoric component of the Cauchy stress:

$$s_1 = J F^{-1} \tau_d F^{-T} \quad \tau_d = \tau + p I.$$
⁽²¹⁾

Further, by letting $\varkappa \to \infty$ Eq. (19) affords a continuous transition from compressible to strictly incompressible behavior (cf. Eq. (16)). Finally, it may be shown that, with a suitable choice of the function w_1 , Eq. (19) reduces to the expression presented by Hermann et al. [8], for near-incompressible linear isotropic elastic materials under small strain.

It is of interest to compare Eq. (16) with a widely quoted expression due to Oden [6]: $\hat{w}_0 = w_0(I_1, I_2) - \lambda(I_3 - 1)/2$. This last expression does not imply that $\lambda = p$ even though $I_1 = J_1$ and $I_2 = J_2$ and $I_3 = J = 1$ a posteriori. For example, let $w_0 = C(I_1 - 3)$. The Cauchy stress τ is

$$\tau = \frac{F\sigma F^{T}}{J} = \frac{2CFF^{T}}{J} - \lambda JI$$
(22)

and hence

$$p = -\frac{\operatorname{trace}\left(\tau\right)}{3} = \lambda - \frac{2CI_1}{3J}.$$
(23)

Equation (19) may be compared with the form proposed by Cescotto and Fonder [7],

$$w = w_1(I_1 - I_3, I_2 - 2I_3) - \lambda(I_3 - 1) - \frac{\lambda^2}{2\varkappa}.$$
(24)

Recalling Eq. (8), it may be shown with some effort that, unlike in the present case, λ in Eq. (24) cannot be equated with p, and w_1 cannot be regarded as a function of the deviatoric Lagrangian strain \bar{e} . Further, as shown in Nicholson [9], the models expressed by Eqs. (22), (24) couple volume and shear even in the small strain limit, and thus do not reduce to the classical thermoelasticity. Also worthy of note are several finite element codes [3], [10] implementing hyperelastic strain energy density functions consistent with Eq. (19).

2.3 Extension to thermal effects: reduced strain

A strain energy function w accommodating thermal expansion is used in the finite element code ABAQUS [3]. Such a function may be sufficient in applications in which the temperature field is known or may be determined independently of the mechanical field. To accommodate thermomechanical effects under more general conditions, such as when thermal and mechanical fields must be determined using coupled equations, we interpret the strain energy density as the Helmholtz free energy density (per unit mass) under isothermal conditions: $w = \varrho \phi$ if $T = T_0$. We now regard ϕ as a function of temperature T and a *reduced* (Cauchy) strain C_R , which is the actual strain C with thermal expansion effects removed (as described below). It possesses invariants I_{1R} , I_{2R} , and $I_{3R} = J_R^2$. Further, we seek a function ϕ having the same structure as its classical thermoelastic counterpart (cf. Appendix A): i.e. $\phi = \phi_M (e_R/J_R) + \phi_C(J_R) + \phi_T(T) + \phi_0$.

The deformation gradient tensor F is expressed in terms of deformations with respect to the initial configuration. The contribution of isotropic thermal expansion is removed as follows:

$$F_R = FF_T^{-1} \quad F_T = \left[1 + \frac{\alpha}{3} (T - T_0)\right] I.$$
 (25)

and the thermal expansion coefficient α is considered constant. Clearly,

$$C_R = f^2(T) C \quad f(T) = \frac{1}{1 + \frac{\alpha}{3} (T - T_0)}.$$
(26)

Suppose the deformation consists of simple isotropic thermal expansion. Then $F = [1 + \alpha(T - T_0)/3] I$. From this it follows that $\varepsilon_R = 0$, $\phi_M = 0$, $\phi_C = 0$, $\sigma = 0$, and p = 0. It is straightforward to show that, under small strain,

$$\varepsilon_R = \frac{1}{2} \left(\boldsymbol{C}_R - \boldsymbol{I} \right) = \boldsymbol{\bar{e}} + \frac{1}{3} \left[\boldsymbol{e} - \boldsymbol{\alpha} (T - T_0) \right] \boldsymbol{I}$$
(27)

where $e = \text{trace}(\varepsilon)$, in agreement with Appendix A. Finally, a simple way to accommodate "small" temperature-shear coupling is described in Appendix B.

2.4 Helmholtz free energy density

Now, if J_{1R} , J_{2R} and J_R^2 denote the invariants of $C_R/J_R^{2/3}$, simple manipulation furnishes

$$J_R = f^3(T) J \quad J_{1R} = J_1 \quad J_{2R} = J_2 \quad K_{2R} = K_2 \quad K_{3R} = K_3.$$
⁽²⁸⁾

Note the important result that $\phi_M(e_R/J_R^{2/3}) = \phi_M(\bar{e}/J^{2/3}) = \phi_M(K_2, K_3)$ so that the term ϕ_M may be regarded as independent of temperature and dilatational effects (represented by J). We assume that, if $\varepsilon = 0$, then $\phi_M = 0$ and $\partial \phi_M/\partial \varepsilon = 0$. Continuing,

$$\phi_C(J_R) = \phi_C(f^3(T) J - 1) \tag{29}$$

so that ϕ_c depends on dilatational effects and temperature. Recalling Eq. (13), the form of Eq. (29) for near-incompressible materials is now

$$\phi_C(J_R) = \frac{\varkappa (f^3(T) J - 1)^2}{2\varrho}.$$
(30)

The final term being introduced is purely temperature dependent. Recall that the entropy η and the specific heat at constant strain c_e are given by

$$\eta = -\frac{\partial \phi}{\partial T} \quad c_e = T \, \frac{\partial \eta}{\partial T}.$$
(31)

Here c_e is not a constant but depends on T and J through ϕ_c . We now write

$$\eta = \eta_C(T, J) + \eta_T(T) \quad \eta_C = -\frac{\partial \phi_C}{\partial T} \quad \eta_T = -\frac{\partial \phi_T}{\partial T}.$$
(32)

For similarity to the linear case (Appendix A), and recalling our assumption that nonlinearity is due primarily to mechanical effects, we make the simple assumption that

$$T \frac{\partial \eta}{\partial T} = \hat{c}_e, \quad \hat{c}_e \text{ known constant.}$$
 (33)

Hereafter the circumflex will not be displayed. Elementary manipulation serves to establish that

$$\eta_T = c_e \ln\left(\frac{T}{T_0}\right). \tag{34}$$

Clearly, other assumptions can be made such as a power series in T can be assumed for η_T .

Combining the foregoing results, the Helmholtz free energy density is

$$\phi = \phi_M(K_2, K_3) + \frac{\varkappa}{2\varrho} \left(f^3(T) J - 1 \right)^2 + c_e T \left[1 - \ln\left(\frac{T}{T_0}\right) \right] + \phi_0.$$
(35)

The 2nd Piola-Kirchhoff stress tensor σ , the entropy density η , and the (true) hydrostatic pressure p are given by

$$\sigma = \rho \,\frac{\partial \phi_M}{\partial \varepsilon} + \varkappa (f^3(T) J - 1) \, f^3(T) \, J C^{-1} \tag{36}$$

$$\eta = \frac{\alpha \varkappa}{\varrho} \left(f^3(T) J - 1 \right) J f^4(T) + c_e \ln\left(\frac{T}{T_0}\right)$$
(37)

$$p = -\frac{1}{3} \operatorname{trace} \left(\tau \right) = -\frac{1}{3J} \operatorname{trace} \left(F \sigma F^T \right) = -\varkappa \left(f^3(T) J - 1 \right) f^3(T).$$
(38)

Earlier expressions for ϕ were proposed by Dillon [1] and Shapery [2], in essence using power series. A very simple expression for ϕ was studied earlier by the present authors [4]. However, it had severe limitations compared to the present expression; for example it did not decouple shear and dilatational effects.

Finally, for computational simulation based on variational principles, for example the finite element method, it may be convenient to write

$$\phi_c = -\frac{p}{\varrho} f^{-3}(T) \left(f^3(T) J - 1 \right) - \frac{p^2}{2\kappa \varrho} f^{-6}(T).$$
(39)

and to assume independent variations of ε and p (and T if thermal equilibrium is considered). Computational aspects of the isothermal counterpart of this expression are discussed in Nicholson [9].

2.5 Applications: thermodynamic stability and the adiabatic tangent modulus matrix

Stability is a concern in modeling behavior of elastomers. For example several finite element codes with hyperelastic elements provide several sample calculations to test for stability [3], [10]. The issue of stability and its implementations for finite element computations will be addressed in a subsequent study. For present purposes, we assume that the material model satisfies criteria for *thermodynamic stability* (Callen [11]).

For later convenience we invoke the VEC operator (cf. Nicholson [9]): Consider a matrix denoted either by a bold upper case letter such as G or its lower case Greek counterpart γ , with entries g_{ij} . The vec operator VEC (G) or VEC (γ) generates the vector g as follows:

$$\boldsymbol{g} = \{g_{11} \ g_{21} \ g_{31} \ g_{12} \ g_{22} \ g_{32} \ g_{13} \ g_{23} \ g_{33}\}^T. \tag{40}$$

The Gibbs free energy $\psi = \varphi - e^T ds/\varrho$ depends on T and s. A sufficient condition for *thermodynamic stability* (Callen [11]) is that the matrix **H** below be positive definite, written H > 0. In particular,

$$d^{2}\psi = -\left\{ dTds \right\} H \begin{cases} dT \\ ds \end{cases}$$
(41)

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where

$$\boldsymbol{H} = \begin{bmatrix} \frac{\partial \eta}{\partial T} \Big|_{\boldsymbol{s}} & \frac{1}{\varrho} & \frac{\partial \varrho^{T}}{\partial T} \Big|_{\boldsymbol{s}} \\ \frac{1}{\varrho} & \frac{\partial \varrho}{\partial T} \Big|_{\boldsymbol{s}} & \frac{1}{\varrho} & \frac{\partial \varrho}{\partial s} \Big|_{\boldsymbol{T}} \end{bmatrix} = \begin{bmatrix} \frac{c_{e'}}{T} + \frac{1}{\varrho} & \beta^{T}\beta & -\frac{\beta^{T}\boldsymbol{D}^{-\frac{1}{2}}}{\varrho} \\ -\frac{\boldsymbol{D}^{-\frac{1}{2}}\beta}{\varrho} & \frac{\boldsymbol{D}^{-1}}{\varrho} \end{bmatrix} = \frac{1}{\varrho} \begin{bmatrix} \boldsymbol{\chi} & \boldsymbol{0}^{T} \\ \boldsymbol{0} & \boldsymbol{D}^{-\frac{1}{2}} \end{bmatrix}^{T} \boldsymbol{H}' \begin{bmatrix} \boldsymbol{\chi} & \boldsymbol{0}^{T} \\ \boldsymbol{0} & \boldsymbol{D}^{-\frac{1}{2}} \end{bmatrix}$$
(42)

in which

$$\boldsymbol{H}' = \begin{bmatrix} 1 & -\frac{\beta^T}{\chi} \\ -\frac{\beta}{\chi} & \boldsymbol{I} \end{bmatrix}$$
(43)

$$\chi = \sqrt{\varrho \, \frac{c_{e'}}{T} + \beta^T \beta} \tag{44}$$

$$\beta = \frac{\boldsymbol{D}^{-\frac{1}{2}} \partial \boldsymbol{s}}{\partial T} \bigg|_{\boldsymbol{e}}$$
(45)

$$c_{e}' = c_{e} + T \frac{\partial \eta_{2}}{\partial T} \bigg|_{e}$$
(46)

$$\boldsymbol{D} = \frac{1}{\varrho} \left. \frac{\partial \boldsymbol{s}}{\partial \boldsymbol{e}} \right|_{T}.$$
(47)

It has been tacitly assumed that the isothermal tangent modulus D is positive definite, and that $\rho c_e'/T + \beta^T \beta > 0$. The following Maxwell relations were also used:

$$\frac{\partial e}{\partial T}\Big|_{\mathbf{s}} = -\mathbf{D}^{-1} \frac{\partial \mathbf{s}^{T}}{\partial T}\Big|_{\mathbf{e}} \quad \frac{\partial \eta}{\partial e}\Big|_{T} = -\frac{1}{\varrho} \frac{\partial \mathbf{s}}{\partial T}\Big|_{\mathbf{e}}.$$
(48)

The condition for thermodynamic stability is satisfied if H' > 0. This may be shown to imply that $\beta^T \beta / \chi^2 < 1$, which is equivalent to $c_{e'} > 0$ and D > 0. Now

$$c_{e}' = c_{e} - \frac{\alpha^{2} \varkappa JT}{3\varrho} f^{5}(T) \left(7f^{3}(T) J - 4\right)$$
(49)

and hence if $c_e' > 0$, c_e must exceed a value depending on α , \varkappa , T and J. For small deviations from the reference configuration and from T_0 , this becomes $c_e > \alpha^2 \varkappa T_0/\varrho$.

The condition D > 0 is commonly assumed in isothermal models, in which D depends only on ε . Here, D also depends on T. We may write $D(\varepsilon, T) = D_1(\varepsilon) + D_2(\varepsilon, T - T_0)$ in which $D(\varepsilon, 0) = D_1(\varepsilon)$. Expressions for D_1 and D_2 based on the work of Nicholson [9] are presented in Appendix C, where the stability conditions are discussed further.

Finally, we note the following expression for the *adiabatic* tangent modulus matrix D_a :

$$\boldsymbol{D}_{a} = \frac{\partial \boldsymbol{s}}{\partial T}\Big|_{\eta} = \boldsymbol{D} + \frac{T}{\varrho c_{e'}} \frac{\partial \boldsymbol{s}}{\partial T}\Big|_{e} \frac{\partial \boldsymbol{s}^{T}}{\partial T}\Big|_{e}.$$
(50)

Recalling that elastomers are often poor conductors of heat, this quantity is especially relevant to applications in which loads are applied rapidly relative to the time required for heat conduction.

3 Application to a rubber rod

The numerical results presented in the subsequent paragraphs assume the following coefficient values (Nicholson and Nelson [4], Gent [12]), which are thought to be representative of unfilled natural rubber:

$$\begin{split} C_1 &= 179 \text{ kN/m}^2 & C_2 &= 15 \text{ kN/m}^2 \\ \varkappa &= 2000 \text{ kN/m}^2 & \alpha &= 2.5 \times 10^{-4} / ^\circ \text{K} \\ c_e &= 1960 \text{ J/kg} - ^\circ \text{K} & \varrho &= 913 \text{ kg/m}^3. \end{split}$$

We now consider three cases involving a rod of the material satisfying

$$\varrho\phi = C_1(J_1 - 3) + (J_2 - 3) + \frac{\varkappa}{2} \left[f^3(T) \left(J - 1 \right) \right]^2 + \varrho c_e T \left[1 - \ln\left(\frac{T}{T_0}\right) \right] + \varrho \phi_0 \tag{51}$$

and

$$d\phi = -\eta dT + \frac{1}{2\varrho} \operatorname{trace} \left(\sigma dC\right) = -\eta dT + \frac{1}{2\varrho} \sum_{i=1}^{3} \sigma_i dc_i$$
(52)

in which $c_i = \lambda_i$ are the principal values of C, λ_i are the principal extension ratios, and σ_i are the principal values of σ . The deformation is isotropic in the transverse plane: $c_2 = c_3$.

It follows that

$$\frac{\sigma_i}{\varrho} = 2 \frac{\partial \phi}{\partial c_i}.$$
(53)

After some manipulation,

$$I_1 = c_1 + 2c_2, \quad I_2 = 2c_1c_2 + c_2^2, \quad I_3 = J^2 = c_1c_2^2.$$
 (54)

Continuing,

$$\sigma_{i} = 2C_{1} \left(1 - \frac{1}{3} \frac{I_{1}}{c_{i}} \right) I_{3}^{-1/3} + 2C_{2} \left(I_{1} - c_{i} - \frac{2}{3} \frac{I_{2}}{c_{i}} \right) I_{3}^{-2/3} + \varkappa f^{3}(T) \left(f^{3}(T) J - 1 \right) \frac{J}{c_{i}}$$
(55)

$$\eta = \frac{\alpha \kappa}{\varrho} \left[f^{3}(T) \left(c_{1} c_{2}^{2} \right)^{1/2} - 1 \right] \left(c_{1} c_{2}^{2} \right)^{1/2} f^{4}(T) + c_{e} \ln \left(\frac{T}{T_{0}} \right).$$
(56)

3.1 Stresses due to temperature increase in a rod of constrained length

Now $c_1 = 1$ and $T = T_0$ (prescribed). The condition $\sigma_2 = 0$ serves to determine c_2 as a function of temperature T. The result may be used to furnish σ_1 as a function of $T - T_0$. The results are displayed in Fig. 1 using $T_0 = 300$ °K.

3.2 Slow isothermal extension of a rubber rod

Now $T = T_0$. The condition $\sigma_2 = 0$ furnishes c_2 as a function of c_1 . Substitution into σ_1 furnishes the stress σ_1 as a function of the strain $\varepsilon_1 = (c_1 - 1)/2$, as shown in Fig. 2.

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3.3 Rapid adiabatic expansion of a rubber rod

Now $\eta = 0$ and $\sigma_2 = 0$. Equations (55) and (56) may be used to obtain c_2 and Tas functions of c_1 . Then Eq. (55) is used to obtain the stress σ_1 as a function of the strain $\varepsilon_1 = (c_1 - 1)/2$ under adiabatic conditions. Assume $T_0 = 300$ °K. The results are shown in Fig. 3.

Appendix A

Classical thermoelasticity

We seek to formulate a thermohyperelastic model retaining the structure of classical thermoelasticity, which we now review. Let ε be the (small) strain tensor, with σ the stress tensor, T the temperature, T_0 the "initial" temperature, and ρ the mass density. The deviatoric strain tensor \bar{e} , the volume strain e, and their counterparts s and s for stress, are given by

$$\bar{e} = \varepsilon - \frac{e}{3}I \quad e = \text{trace}(\varepsilon)$$

$$s = \sigma - \frac{s}{3}I \quad s = \text{trace}(\sigma)$$
(A.1)

where I is the identity tensor. The constitutive relations of interest are

$$s = 2\mu\bar{e} \quad s = 3\varkappa[e - \alpha(T - T_0)] \quad T \frac{\partial\eta}{\partial T} = c_e.$$
(A.2)

Here μ is the shear modulus, \varkappa is the bulk modulus, c_e is the specific heat at constant strain, and α is the volumetric coefficient of thermal expansion, all assumed to be constants. Note the use of the reduced volume strain $e_R = e - \alpha (T - T_0)$, which is the actual volume strain reduced by the extent of thermal expansion. The isotropic stress s and the hydrostatic pressure p = -s/3 are functions of e_R .

Thermodynamically, the stress tensor σ and the entropy density η are obtained from the Helmholtz free energy density using

$$d\phi = -\eta dT + \frac{1}{\varrho} \operatorname{trace} (\sigma d\varepsilon). \tag{A.3}$$

Application of the foregoing constitutive relations furnishes

$$\phi = \phi_M + \phi_C + \phi_T + \phi_0$$

$$\phi_M = \frac{\mu}{\varrho} \operatorname{trace} \left(\bar{e}^2\right) \quad \phi_C = \frac{\kappa}{2\varrho} e^2 - \frac{\varkappa \alpha e}{\varrho} \left(T - T_0\right)$$

$$\phi_T = c_e T \left[1 - \ln\left(\frac{T}{T_0}\right)\right] \quad \eta = c_e \ln\left(\frac{T}{T_0}\right) + \frac{\varkappa \alpha e}{\varrho}.$$
(A.4)

Clearly, ϕ_M represents "shear" and is independent of temperature *T* and the volume strain *e*. Similarly, ϕ_T is a pure thermal term, and is not dependent on strain. The remaining term ϕ_C "couples" the volume strain *e* to the temperature *T*. Finally, ϕ_0 represents the value of ϕ in the reference configuration.

Appendix B

Treatment of "small" temperature-shear coupling

We expect that the primary effect of temperature takes the form of thermal expansion. However, it seems quite reasonable to consider that there may be a "small" (compared to the volume) effect evident in the "shear" response. A simple way to consider such an effect would be to modify the function $\phi_M(K_2, K_3)$ as follows:

$$\phi_{M}(K_{2}, K_{3}, T) = \phi_{M}(K_{2}, K_{3}) \left[1 + \gamma(T - T_{0})\right].$$
(B.1)

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Equations (36) and (37) now become

$$\sigma = \varrho [1 - \gamma (T - T_0)] \frac{\partial \phi_M}{\partial \varepsilon} + \varkappa (f^3(T) J - 1) f^3(T) J C^{-1}$$
(B.2)

$$\eta = \alpha \varkappa \left(f^{3}(T) J - 1 \right) J f^{4}(T) + c_{e} \ln \left(\frac{T}{T_{0}} \right) - \gamma \phi_{M}(K_{2}, K_{3}).$$
(B.3)

Appendix C

Temperature-dependent tangent modulus matrix

Relations reported by Nicholson [9] under isothermal conditions are readily extended to the present model to furnish

$$D = 2 \sum_{i=1}^{3} \sum_{j=1}^{3} \phi_{ij} n_i n_j^T + 2 \sum_{i=1}^{3} \phi_i A_i \quad c = \text{VEC}(C) \quad c_2 = \text{VEC}(C^2)$$
(C.1)

$$\boldsymbol{n}_i^T = \frac{\partial I_i}{\partial c} \quad \phi_{ij} = \frac{\partial^2 \phi}{\partial I_i \partial I_j} \quad \phi_i = \frac{\partial \phi}{\partial I_i} \quad \boldsymbol{A}_i = \frac{\partial \boldsymbol{n}_i}{\partial c}. \tag{C.2}$$

Further,

$$n_{i} = i \quad n_{2} = I_{1}i - c \quad n_{3} = c_{2} - I_{1}c + I_{2}i$$

$$A_{1} = 0 \quad A_{2} = ii^{T} - I_{9} \quad A_{3} = I \otimes C + C \otimes I - (ic^{T} + ci^{T}) + I_{1}(ii^{T} - I_{9})$$
(C.3)

in which \otimes denotes the Kronecker product of two matrices.

$$\varrho\phi = C_1(J_1 - 3) + C_2(J_2 - 3) + \frac{\varkappa}{2} \left(f^3(T) J - 1 \right)^2 + \varrho c_e T \left[1 - \ln\left(\frac{T}{T_0}\right) \right] + \varrho \phi_0.$$
(C.4)

We apply these relation to the near-incompressible counterpart of the two-term incompressible Mooney-Rivlin material. It follows that

$$\phi_{1} = C_{1}I_{3}^{-\frac{1}{3}} \qquad \phi_{2} = C_{2}I_{3}^{-\frac{2}{3}}$$

$$\phi_{3} = \frac{\kappa f^{3}(T)}{2J} \left(f^{3}(T)J - 1 \right) - \frac{1}{3}C_{1}I_{1}I_{3}^{-\frac{4}{3}} - \frac{2}{3}C_{2}I_{2}I_{3}^{-\frac{5}{3}}.$$
(C.5)

Thus, for the assumed material,

$$\phi_{11} = \phi_{22} = \phi_{12} = \phi_{21} = \phi_{1T} = \phi_{2T} = 0$$

$$\phi_{31} = \phi_{13} = -\frac{1}{3} C_1 I_3^{-\frac{4}{3}}$$

$$\phi_{32} = \phi_{23} = -\frac{2}{3} C_2 I_3^{-\frac{5}{3}}$$

$$\phi_{33} = \frac{\varkappa f^3(T)}{4} I_3^{-\frac{3}{2}} + \frac{4}{9} C_1 I_1 I_3^{-\frac{7}{3}} + \frac{10}{9} C_2 I_2 I_3^{-\frac{8}{3}}$$

$$\phi_{3T} = -\frac{2\alpha\varkappa}{3} f^7(T) + \frac{\alpha\varkappa}{2J} f^4(T).$$

(C.6)

We now write $f^{3}(T) = 1 + g_{3}(T)$ and $f^{6}(T) = 1 + g_{6}(T)$, and note that $g_{3}(0) = g_{6}(0) = 0$. Some manipulation serves to derive the following compact expressions:

$$D_{1}(\varepsilon) = 2C_{2}A_{2}I_{3}^{-\frac{2}{3}} + \left[\varkappa\left(1 - \frac{1}{J}\right) - \frac{2}{3}C_{1}I_{1}I_{3}^{-\frac{4}{3}} - \frac{4}{3}C_{2}I_{2}I_{3}^{-\frac{5}{3}}\right]A_{3} - \frac{2}{3}C_{1}I_{3}^{-\frac{4}{3}}(\boldsymbol{n}_{1}\boldsymbol{n}_{3}^{T} + \boldsymbol{n}_{3}\boldsymbol{n}_{1}^{T}) - \frac{4}{3}C_{2}I_{3}^{-\frac{5}{3}}(\boldsymbol{n}_{2}\boldsymbol{n}_{3}^{T} + \boldsymbol{n}_{3}\boldsymbol{n}_{2}^{T}) + \left[\frac{\varkappa}{2}I_{3}^{-\frac{3}{2}} + \frac{8}{9}C_{1}I_{1}I_{3}^{-\frac{7}{3}} + \frac{20}{9}C_{2}I_{2}I_{3}^{-\frac{8}{3}}\right]\boldsymbol{n}_{3}\boldsymbol{n}_{3}^{T}$$
(C.7)

$$\boldsymbol{D}_{2}(\varepsilon, T-T_{0}) = \varkappa \left(g_{6}(T) - \frac{g_{3}(T)}{J} \right) A_{3} + \frac{\varkappa}{2} g_{3}(T) I_{3}^{-\frac{3}{2}} \boldsymbol{n}_{3} \boldsymbol{n}_{3}^{T}.$$
(C.8)

Presumably, when $T = T_0$, at strain ε the stability condition $D = D_1 > 0$ is satisfied implying restrictions on C_1 , C_2 , and \varkappa . Now suppose $T \neq T_0$. The condition $D_1 + D_2 > 0$ now imposes restrictions on α as well as on C_1 , C_2 , and \varkappa . Suppose that, based on test data or other information, the values C_1^* , C_2^* , \varkappa^* and α^* are estimated for a given elastomer. The foregoing relations can be used to furnish a computational assessment of the range of ε and T over which the stability conditions are satisfied.

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