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ORIGINAL ARTICLE

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Hybrid free energy approach for nearly incompressible behaviors at finite strain

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Abstract We explore the formulation of nearly incompressible behaviors at finite strain in the context of a hybrid or a mixed energy. Such an energy is a function of both an isochoric deformation and a pressure-like quantity that can be considered as an internal variable. From thermodynamical and physical considerations, new energy functions are developed to correctly describe both nearly incompressible elasticity and thermoelastic behaviors. We discuss the advantages of such a formulation; in particular, we show that this approach makes it possible to unify the variational and the thermodynamical formulations in the nearly incompressible context without using Lagrange multipliers or other specific variational principles.

Keywords Thermomechanical coupling · Entropic elasticity · Free energy · Volumetric behavior

1 Introduction

In the literature, the isochoric/volumetric decomposition, as earlier proposed by [8], is widely used to take into account nearly incompressible behaviors at finite strain. Near incompressiblity, the levels of energy associated with an isochoric or a volumetric transformation are clearly not of the same order of magnitude, which allows a split of the strain energy. Flory has proposed a multiplicative split of the deformation gradient into a purely volumetric part and a purely isochoric part. Standard formulations based on the Helmholtz free energy lead to consider that the thermodynamic potentials depend on these strain variables: isochoric and volumetric gradients (e.g., [11,19,21,25]). An alternative to these strain-based approaches would be to consider the Gibbs formulation for which thermodynamic potentials depend on stress quantities (e.g., [13,17]). The Helmholtz approach is widely used by the mechanical community, while Gibbs is more popular in the chemical community. A strain (or a stress) decomposition does not imply the introduction of an additional thermodynamic variable, but it makes it possible to separately build the isochoric and the volumetric parts of the strain energy.

From a numerical point of view, the variational formulation for nearly incompressible problems requires specific treatments such as Lagrange multipliers (within multi-field variational principles), or projection methods or reduced/selective integration strategies to avoid locking phenomena. The pioneer work of Reissner has opened up a field of research on the development of variational principles that could take into account a volume constraint in the nonlinear framework (c.f. [1,22]). The developments of Reissner and co-workers were based on a partial Legendre transformation of the strain energy which is split into volumetric and isochoric parts.

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They showed that a two-field form based on the Hellinger–Reissner principle can be explicit in the small strain range, and the generalization to the nonlinear case cannot be explicit in general. An alternative to these two-field methods is to use a three-field form such as the Hu–Washizu principle (e.g., [24] for an application to finite strain elasto-plasticity). However, all these methods consider a standard Helmholtz formulation for the thermodynamic framework and the variable transformation is only introduced in the weak form. In this sense, thermodynamical and variational formulations may differ for nearly incompressible media. Numerical problems may occur in the context of coupled thermomechanical behaviors where the thermoelastic coupling contribution is linked to the volumetric behavior.

As an alternative to the Gibbs or Helmholtz approaches, some authors have proposed to consider a partial Legendre transformation of free energy within the thermodynamic framework. In [16], the authors propose a mixed or hybrid free energy in which the hydrostatic pressure is introduced as an alternative to the volume variation. The isochoric part of the free energy remains unchanged, and they obtain a mixed formulation with both strain and stress variables. This formulation is interesting when analyzing the calorimetric behavior of nearly incompressible materials because the experiments are done at constant pressure rather than at constant volume. As shown in [15,16], this hybrid free energy approach is also very interesting in the context of materials undergoing chemical reactions, because the hydrostatic pressure is a natural variable for modeling chemical processes. Moreover, a mixed or hybrid free energy approach makes it possible to obtain a multi-field variational formulation equivalent in some cases to a Hellinger–Reissner formulation.

In this work, we propose a general formulation of hybrid free energies. We do not restrict ourselves to the case of a standard partial Legendre transform of a volumetric free energy formulated from the variation of volume. An additional internal scalar variable (pressure-like variable) is introduced in the formulation. This variable is related to the hydrostatic pressure through the definition of a volumetric energy which depends on this internal variable and if needed on the temperature. This is a more flexible approach that allows us to take into account a volume viscosity or volume thermoelastic effects. Thermodynamic and physical considerations are also discussed to obtain well-defined potentials.

This paper is structured as follows. In Sect. 2, the fundamental thermodynamic framework is recalled and the concept of hybrid energy is introduced. In Sect. 3, two hybrid energy functions are proposed and discussed in the case of nearly incompressible isothermal elasticity. For the first energy function proposed, we show that we can explicitly recover a volumetric function previously proposed by [18]. For the second energy function, we cannot explicitly formulate the inverse function in terms of volume. On a simple extension test, we discuss, for both functions, the unphysical behavior that could occur depending on the interplay between isochoric and volumetric or hydrostatic parts (see [6,10]). In Sect. 4, we show that the pressure-like variable can be used to model inelastic and irreversible (viscosity in this paper) volume effects. In Sect. 5, we consider the case of entropic elasticity (e.g., see [12]). We discuss the effect of volumetric thermoelastic coupling for behaviors where the volumetric thermal expansion is taken into account.

2 Thermodynamics and hybrid free energy

In the following, we adopt a standard thermodynamic framework of continuous media. We assume that irreversibilities are described by internal variables, i.e., we assume that the so-called principle of local action holds. The initial stress-free configuration (or reference configuration) is called Ω_0. The current configuration is $\Omega(t)$. A material point $\mathbf{X}$ in the initial configuration is bijectively transformed into a point $\mathbf{x}(\mathbf{X}, t)$ in the current configuration. The gradient of the transformation is $\mathbf{F}(\mathbf{X}, t)$, and $J(\mathbf{X}, t) = \det\mathbf{F}(\mathbf{X}, t)$ is the change in volume. The current local density, $\rho(\mathbf{X}, t)$, and the reference one, $\rho_0(\mathbf{X})$, are related together with the conservation of mass for this closed system: $\rho = J\rho_0$.

2.1 Hybrid free energy

Let ψ be the specific Helmholtz free energy. We assume that this energy depends on the isochoric deformation gradient $\bar{\mathbf{F}} = (J^{-1/3}\mathbf{1})\mathbf{F}$, the temperature Θ, the volume variation J and n internal variables α_i, if needed:

$$\psi = \hat{\psi}(\bar{\mathbf{F}}, \Theta, J, \alpha_i) \qquad i = 1 \ldots n \tag{1}$$

This formulation has been widely adopted because of the isochoric/ volumetric split previously proposed by [8]: the energy $\hat{\psi}$ can be split into an isochoric and a volumetric part. Different volumetric potentials can be found in the literature (see [10] for a discussion about volumetric potentials).

The key feature of the hybrid free energy formulation consists in considering the following hybrid (or mixed) free energy:

$$\boxed{\varphi(\bar{\mathbf{F}}, \Theta, q, \alpha_i) = \hat{\psi}(\bar{\mathbf{F}}, \Theta, J, \alpha_i) + \beta(J, \Theta, q) \qquad i = 1 \dots n} \tag{2}$$

where q is a scalar pressure-like variable that can be considered as an additional internal variable. We therefore suppose that the evolution of q is independent of the evolution of other state and internal variables and we deduce from thermodynamic principles a constitutive equation for the variable q. The function β, which will be discussed later in detail, plays a central role in our approach. It aims at liking on the one side the pressure-like variable q and the volume variation J and, on the other side, the hydrostatic pressure p and the stress-like variable q. We use the term “hybrid” free energy as originally proposed by [16], and we then show that the function β can be chosen such that we recover a partial Legendre transformation of the standard Helmholtz energy and the stress variable q is therefore equal to the hydrostatic pressure. Hybrid free energy can be related to internal energy, e, and entropy, s, using the relation $\hat{\psi} = e - s\Theta$, we obtain:

$$\boxed{\varphi = e - s\Theta + \beta} \tag{3}$$

2.2 Thermodynamics principles and Clausius–Duhem inequality

The conservation of energy (first principle of thermodynamics) in the reference configuration can be written in a local form as:

$$\rho_0 \dot{e} = \mathbf{\Pi} : \dot{\mathbf{F}} + \rho_0 r - \mathrm{div}_X \mathbf{Q}_\Theta \tag{4}$$

where $\mathbf{\Pi}$ is the first Piola–Kirchoff stress, r is an external heat source, div_X is the Lagrangian divergence and $\mathbf{Q}_\Theta$ is the Lagrangian heat flux. The entropy production (second principle of thermodynamics) in the reference configuration can be written under the following local form:

$$\rho_0 \Theta \dot{s} - \rho_0 r + \mathrm{div}_X \mathbf{Q}_\Theta - \frac{\mathrm{grad}_{\mathbf{X}} \Theta}{\Theta} \cdot \mathbf{Q}_\Theta \geq 0 \tag{5}$$

where $\mathrm{grad}_{\mathbf{X}}$ is the gradient in the reference configuration. Differentiating Eq. (3) with respect to time, one has:

$$\dot{\varphi} = \dot{e} - \dot{s}\Theta - s\dot{\Theta} + \dot{\beta} \tag{6}$$

and combining Eqs. (6) and (4) in Eq. (5), the so-called Clausius–Duhem inequality is obtained:

$$\phi = \mathbf{\Pi} : \dot{\mathbf{F}} - \rho_0 \dot{\varphi} + \rho_0 \dot{\beta} - \rho_0 s \dot{\Theta} - \frac{\mathrm{grad}_{\mathbf{X}} \Theta}{\Theta} \cdot \mathbf{Q}_\Theta \geq 0 \quad \forall \dot{\mathbf{F}}, \dot{\Theta}, \mathbf{Q}_\Theta \tag{7}$$

where ϕ is the dissipation. Using the definition of Eq. 2, the time derivative of the hybrid free energy and the volumic energy can be computed such that:

$$\dot{\varphi} = \frac{\partial \varphi}{\partial \bar{\mathbf{F}}} : \dot{\bar{\mathbf{F}}} + \frac{\partial \varphi}{\partial \alpha_i} \bullet \alpha_i + \frac{\partial \varphi}{\partial q} \dot{q} + \frac{\partial \varphi}{\partial \Theta} \dot{\Theta} \tag{8}$$

$$\dot{\beta} = \frac{\partial \beta}{\partial J} \dot{J} + \frac{\partial \beta}{\partial q} \dot{q} + \frac{\partial \beta}{\partial \Theta} \dot{\Theta} \tag{9}$$

where $\bullet$ is an operator depending on the internal variables α_i such that the product $\partial\varphi/\partial\alpha_i \bullet \alpha_i$ leads to a scalar quantity. The time derivative of strain quantities leads to the following expressions:

$$\dot{J} = J \mathbf{F}^{-T} : \dot{\mathbf{F}} \tag{10}$$

$$\dot{\bar{\mathbf{F}}} = J^{-1/3} \left[\mathbb{I} - \frac{1}{3} \mathbf{F} \otimes \mathbf{F}^{-T} \right] : \dot{\mathbf{F}} = \mathbb{P}_{\mathbf{F}} : \dot{\mathbf{F}} \tag{11}$$

where $\mathbb{I}$ is the fourth-order identity tensor. Inserting Eqs. (10), (11) in Eqs. (8) and (9) and combining with Eq. (7), the Clausius–Duhem inequality can be rewritten as follows:

$$\boxed{\begin{aligned}\phi = &\left(\boldsymbol{\Pi} - \rho_0 \frac{\partial \varphi}{\partial \bar{\mathbf{F}}} : \mathbb{P}_{\mathbf{F}} + \rho_0 J \frac{\partial \beta}{\partial J} \mathbf{F}^{-T}\right) : \dot{\mathbf{F}} - \rho_0 \left(s + \frac{\partial \varphi}{\partial \Theta} - \frac{\partial \beta}{\partial \Theta}\right) \dot{\Theta} \\ &- \rho_0 \frac{\partial \varphi}{\partial \alpha_i} \bullet \dot{\alpha}_i - \rho_0 \left(\frac{\partial \varphi}{\partial q} - \frac{\partial \beta}{\partial q}\right) \dot{q} - \frac{\operatorname{grad}_X \Theta}{\Theta} \cdot \mathbf{Q}_\Theta \geq 0 \qquad \forall \mathbf{D}, \dot{\alpha}_i, \dot{\Theta}, \dot{q}, \mathbf{Q}_\Theta\end{aligned}} \tag{12}$$

We admit that the dissipation separates into intrinsic dissipation and thermal dissipation, which are assumed to be independently positive, and we assume that the Helmholtz postulate holds, i.e., we can impose thermal variations to a thermodynamic system that remains in its mechanical equilibrium. We therefore obtain a relation between entropy and hybrid energy,

$$s = -\frac{\partial \varphi}{\partial \Theta} + \frac{\partial \beta}{\partial \Theta} \tag{13}$$

and the following expressions for the dissipation terms:

$$\begin{aligned}\phi^{\text{int}} = &\left(\boldsymbol{\Pi} - \rho_0 \frac{\partial \varphi}{\partial \bar{\mathbf{F}}} : \mathbb{P}_{\mathbf{F}} + \rho_0 J \frac{\partial \beta}{\partial J} \mathbf{F}^{-T}\right) : \dot{\mathbf{F}} - \rho_0 \frac{\partial \varphi}{\partial \alpha_i} \bullet \dot{\alpha}_i \\ &- \rho_0 \left(\frac{\partial \varphi}{\partial q} - \frac{\partial \beta}{\partial q}\right) \dot{q} \geq 0\end{aligned} \tag{14}$$

$$\phi^{\text{therm}} = -\frac{\operatorname{grad}_X \Theta}{\Theta} \cdot \mathbf{Q}_\Theta \geq 0 \tag{15}$$

In contrast to the case of a standard Helmholtz formulation, it can be seen from Eq. (14) that we need to define an additional equation related to $\dot{q}$. This can be done, assuming standard generalized materials and defining a supplementary thermodynamic function: the so-called pseudo-potential of dissipation (see [9]). In the following, we adopt an alternative approach.

2.3 Some restrictions on the hybrid free energy

As with the standard Helmholtz free energy, we consider the following restrictions for the stress variable q in the case of nearly incompressible behaviors:

- limit conditions: if we restrict ourselves to undamaged cases, the hybrid energy should tend to infinity when the stress-like variable q tends to infinity (positively or negatively).
- normality condition: we can assume that in the initial configuration $q = 0$ and $\varphi = 0$ hold.
- convexity conditions: in the elastic and isothermal case, we can consider a convexity condition for $|\varphi|$ respective to the variable q independently of each other so as to be in position to satisfy the polyconvexity condition of Ball (see, e.g., [2,5]). The function φ can be convex or concave on q depending on the choice made for function β (the function φ can take positive or negative values). This will be discussed in the next section.

3 Nearly incompressible isothermal elasticity

In this section, we consider the case of elastic materials under isothermal (adiabatic) conditions. In this case, the free energy does not depend on internal variables and the Clausius–Duhem inequality is reduced to the intrinsic dissipation, Eq. (14), which vanishes in this case:

$$\phi^{\text{int}} = \left(\boldsymbol{\Pi} - \rho_0 \frac{\partial \varphi}{\partial \bar{\mathbf{F}}} : \mathbb{P}_{\mathbf{F}} + \rho_0 J \frac{\partial \beta}{\partial J} \mathbf{F}^{-T}\right) : \dot{\mathbf{F}} - \rho_0 \left(\frac{\partial \varphi}{\partial q} - \frac{\partial \beta}{\partial q}\right) \dot{q} = 0 \quad \forall \dot{\mathbf{F}}, \dot{q} \tag{16}$$

From the previous equation, we obtain two constitutive laws, one for the stress and the other one for the compressibility relation:

$$\boldsymbol{\Pi} = \bar{\boldsymbol{\Pi}} : \mathbb{P}_{\mathbf{F}} - \rho_0 J \frac{\partial \beta}{\partial J} \mathbf{F}^{-T} \tag{17}$$

$$\frac{\partial \varphi}{\partial q} = \frac{\partial \beta}{\partial q} \tag{18}$$

where the stress $\bar{\mathbf{\Pi}}$ is defined from: $\bar{\mathbf{\Pi}} = \rho_0 \partial\varphi/\partial\bar{\mathbf{F}}$. It can be seen that the hydrostatic pressure is defined by the variation of the potential beta (and is therefore related to q):

$$p = -\rho_0 \frac{\partial \beta}{\partial J} \tag{19}$$

3.1 Isochoric/volumetric split of the hybrid free energy

As in the case of nearly incompressible elasticity, we consider the isochoric/volumetric split, as earlier proposed by [7]. We assume that the hybrid energy contribution related to an equilibrium state where only the stress variable q varies is much stronger than that involving only isochoric deformation. It seems reasonable to consider the following split:

$$\varphi(\bar{\mathbf{F}}, q) = \varphi_{iso}(\bar{\mathbf{F}}) + \varphi_{\mathrm{vol}}(q) = \varphi_{iso}(\bar{\mathbf{F}}) - \frac{g(q)}{\rho_0} \tag{20}$$

where g is a scalar function of q (defined on $\mathbb{R}$).

Equation (19) shows that the variation of β with respect to J should depend on q, and eventually on other state variables, to define a relation between q and the hydrostatic pressure p. Therefore, we propose the following multiplicative form:

$$\beta(J, q) = q\frac{h(J)}{\rho_0} \tag{21}$$

It can be noticed that $\varphi_{iso}(\bar{\mathbf{F}}) = \hat{\psi}_{iso}(\bar{\mathbf{F}})$ and:

$$\hat{\psi}_{\mathrm{vol}}(J) = q\frac{h(J)}{\rho_0} - \frac{g(q)}{\rho_0} \tag{22}$$

therefore, the function $g(q)$ can be viewed as a Legendre–Flenchel transformation of $\hat{\psi}_{\mathrm{vol}}(J)$. As mentioned previously, q and J are dual variables, and if g is convex, $\hat{\psi}_{\mathrm{vol}}(J)$ will also be convex. Therefore, restricting ourselves to only convex functions for g, and assuming that $\varphi_{iso}(\bar{\mathbf{F}})$ satisfies polyconvexity and coercivity conditions, the existence theorem of Ball could be satisfied if coerciveness is also satisfied (e.g., see [2]).

The stress-like variable q is related to p and J through Eqs. (19) and (18):

$$p = -q h'(J) \qquad \text{and} \qquad h(J) = -g'(q) \tag{23}$$

A straightforward choice is $h(J) = 1 - J$ because it leads to the equality $p = q$. We can also notice that in this case the volumetric part of the hybrid free energy is equivalent to the volumetric part of the free enthalpy as already shown in [16].

For g, we propose to study two different cases:

$$g_1(q) = \frac{k}{a^2}\left(\exp\left(\frac{q}{k}\right)\right)^a - \frac{q}{a} - \frac{k}{a^2} \tag{24}$$

$$g_2(q) = \frac{b(1+c)k}{c}\log\left(1 + c\exp\left(\frac{q}{k}\right)\right) - bq - b\frac{k}{c}(1+c)\log(1+c) \tag{25}$$

where k is a bulk-like modulus (dimension of a stress) and $a \geq 1$, $0 < b \leq 1$ and $c \geq 0$ are dimensionless parameters. Both functions are convex and fulfill the condition $g(0) = 0$. Figure 1(a) illustrates the two functions for different values of a, b and c. Using Eq. (23) leads to the following pressure/volume constitutive relations:

$$(J-1) = \frac{1}{a}\left(\left(\exp\left(\frac{q}{k}\right)\right)^a - 1\right) \tag{26}$$

$$(J-1) = b\frac{\exp\left(\frac{q}{k}\right) - 1}{c\exp\left(\frac{q}{k}\right) + 1} \tag{27}$$

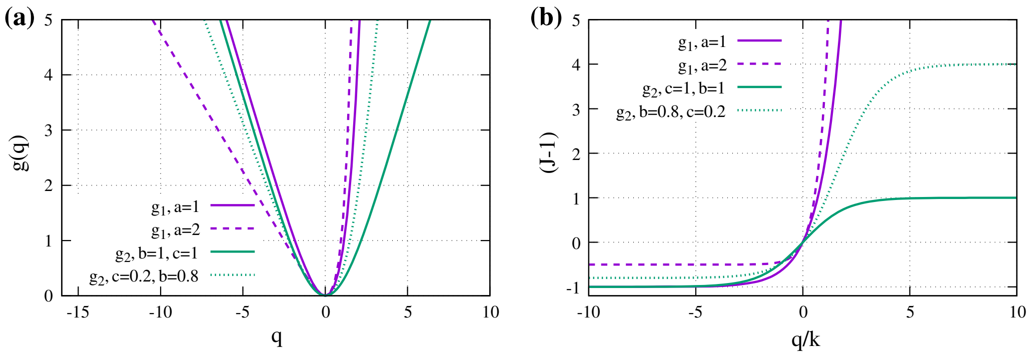

Fig. 1 Volumetric contribution for the two models g_1 and g_2 for different values of a. **a** Volumetric hybrid energy (for $k = 1\ MPa$). **b** Volume variation vs normalized hydrostatic pressure

The choice of the two functions g_1 and g_2 was originally motivated by the integration of Eqs. (26) and (27) in the case $a = b = c = 1$. We have chosen an exponential form in order to have $q \to -\infty$ if $J \to 0$ (obviously $J \leq 0$ is a non- admissible domain). We will show latter that choosing $a = 1$ or $b = c = 1$ may lead in some cases to unphysical responses.

Figure 1b illustrates pressure/volume relations of Eqs. (26) and (27) for different values of a, b and c. The parameters a, b and c can be considered as compressibility limits: as in the statistical theory of rubber elasticity where a limit of extensibility of polymeric chains is taken into account, we investigate the case where the volumetric behavior may exhibit such limits, i.e., the case where $\lim_{J \to J_l} \varphi \to \infty$ (J_l is a compressibility limit in hydrostatic tension or compression). In the case of the first model, only the hydrostatic compression behavior may have a compressibility limit (as an alternative to the physical restriction $J > 0$) such as $J_l = 1 - 1/a$. With this model, the hydrostatic tension leads to an infinite volume variation when $q \to \infty$. With the second model, the compressibility limit in hydrostatic compression is $J_l = 1 - b$ and $J_l = 1 + b/c$ for hydrostatic tension.

Remark 1 The linearization near $q = 0$ of both volumetric relations proposed Eqs. (26) and (27) leads to:

$$(J - 1) = \frac{q}{k} + O(q^2) \tag{28}$$

$$(J - 1) = \frac{b}{1 + c}\frac{q}{k} + O(q^2) \tag{29}$$

For the first model, the parameter k is equivalent to the infinitesimal compressibility modulus. For the second model, the infinitesimal compressibility modulus is equal to $(1 + c)k/b$.

Remark 2 The relation of Eq. (26) can be explicitly inverted: $q = k \log((a(J - 1) + 1)^{-a})$; for $a = 1$, we recover the pressure law of [18]: $q = k \log(J)$. The function g_1, with $a = 1$, is an explicit Legendre transformation of the free energy of [18]. Another simple example of a partial Legendre transformation is to consider the function $g(q) = q^2/(2k)$. If $h(J) = 1 - J$, we obtain the linear flow rule $q = p = k(J - 1)$; however, this relation does clearly not fulfill the physical requirement $J > 0$ independently of q.

Remark 3 If we choose another relation for h as for instance $h(J) = -\ln(J)$, the stress variable q may not be strictly equal to the hydrostatic pressure. In this case, we obtain $p = q/J$.

3.2 Tangent moduli

We consider the tangent moduli defined from the variation of the first Piola–Kirchoff stress in Eq. (38), such that:

$$\begin{aligned} \delta \mathbf{\Pi} &= \mathbb{C}_{\mathbf{FF}} : \delta \mathbf{F} + \mathbb{C}_{\mathbf{F}q} \delta q \\ &= \delta(\bar{\mathbf{\Pi}} : \mathbb{P}_{\mathbf{F}}) - \left(Jq(h' + Jh'')\mathbf{F}^{\text{-T}} \otimes \mathbf{F}^{\text{-T}} - Jqh'\mathbf{F}^{\text{-T}} \oslash \mathbf{F}^{\text{-T}}\right) : \delta \mathbf{F} - (Jh'\mathbf{F}^{\text{-T}})\delta q \end{aligned} \tag{30}$$

where $\obslash$ is a tensor operator such that: $\mathbf{A} \obslash \mathbf{B} = A_{il} B_{kj}$. After some algebraic manipulations, the variation of the first term can be obtained:

$$\begin{aligned}\delta(\bar{\mathbf{\Pi}} : \mathbb{P}_{\mathbf{F}}) = &\left((\mathbb{P}_{\mathbf{F}}^{\mathrm{T}} : \bar{\mathbf{\Pi}} : \mathbb{P}_{\mathbf{F}}) - \frac{J^{-1/3}}{3} (\bar{\mathbf{\Pi}} \otimes \mathbf{F}^{-\mathrm{T}} + \mathbf{F}^{-\mathrm{T}} \otimes \bar{\mathbf{\Pi}}) \right) : \delta\mathbf{F} \\ &+ \left(\frac{J^{-1/3}}{3} (\bar{\mathbf{\Pi}} : \mathbf{F})(\mathbf{F}^{-\mathrm{T}} \obslash \mathbf{F}^{-\mathrm{T}} + \frac{1}{3} \mathbf{F}^{-\mathrm{T}} \otimes \mathbf{F}^{-\mathrm{T}}) \right) : \delta\mathbf{F}\end{aligned} \tag{31}$$

Combining Eqs. (30) and (31), we obtain $\mathbb{C}_{\mathbf{FF}}$:

$$\begin{aligned}\mathbb{C}_{\mathbf{FF}} = &(\mathbb{P}_{\mathbf{F}}^{\mathrm{T}} : \bar{\mathbf{\Pi}} : \mathbb{P}_{\mathbf{F}}) - \frac{J^{-1/3}}{3} (\bar{\mathbf{\Pi}} \otimes \mathbf{F}^{-\mathrm{T}} + \mathbf{F}^{-\mathrm{T}} \otimes \bar{\mathbf{\Pi}}) \\ &+ \frac{J^{-1/3}}{3} (\bar{\mathbf{\Pi}} : \mathbf{F})(\mathbf{F}^{-\mathrm{T}} \obslash \mathbf{F}^{-\mathrm{T}} + \frac{1}{3} \mathbf{F}^{-\mathrm{T}} \otimes \mathbf{F}^{-\mathrm{T}}) \\ &+ \big(Jq(h' + Jh'') \mathbf{F}^{\text{-T}} \otimes \mathbf{F}^{\text{-T}} - Jqh' \mathbf{F}^{\text{-T}} \obslash \mathbf{F}^{\text{-T}} \big)\end{aligned} \tag{32}$$

This set of equations can be completed by the variation of the pressure law:

$$-h'(J) J \mathbf{F}^{-\mathrm{T}} : \delta\mathbf{F} + g'' \delta q = \mathbb{C}_{q\mathbf{F}} : \delta\mathbf{F} + \mathbb{C}_{qq} \delta q = 0 \tag{33}$$

We can observe that $\mathbb{C}_{\mathbf{F}q} = \mathbb{C}_{q\mathbf{F}}$ regardless of the choice we make for the functions $h(J)$ and $g(q)$. We can also notice that the hybrid free energy allows for a straightforward mixed variational formulation (u, q) without considering a Lagrangian multiplier technique or a specific variational principle. An advantage of this formulation is that we obtain a symmetric tangent operator regardless of the choice made for the volumetric functions h and g.

3.3 Free extension case

We restrict ourselves to the case of isotropic elasticity. We investigate the case of a simple compressible medium in extension for a given deformation gradient defined as: $\mathbf{F} = \lambda \vec{e}_1 \otimes \vec{e}_1 + \sqrt{J/\lambda}\, \vec{e}_2 \otimes \vec{e}_2 + \sqrt{J/\lambda}\, \vec{e}_3 \otimes \vec{e}_3$. In [6], the authors showed that the volumetric/isochoric split may lead to an unphysical response for a simple extension (compressible case). In [10], this unphysical response is discussed. The authors showed that this is mainly related to the interplay between the isochoric and the volumetric strain-energy parts.

In this section, we adopt a simple neo-Hookean isochoric hybrid energy: $\varphi_{iso} = c_{10}(\bar{I}_1 - 3)$ where $\bar{I}_1 = \mathrm{tr}(\bar{\mathbf{C}})$ with $\bar{\mathbf{C}} = \bar{\mathbf{F}}^{\mathrm{T}} \bar{\mathbf{F}}$. The functions h and g are chosen to be the same as the one discussed in Sect. 3.1. The stress and the hydrostatic pressure are given by:

$$\mathbf{\Pi} = 2c_{10} J^{-2/3} (\mathbf{F} - \tfrac{I_1}{3} \mathbf{F}^{-\mathrm{T}}) + pJ\mathbf{F}^{-\mathrm{T}} \tag{34}$$

$$(J-1) = g_1'(p) \quad \text{or} \quad (J-1) = g_2'(p) \tag{35}$$

The hydrostatic pressure can be eliminated from the condition $\mathbf{\Pi}_{22} = 0$, and thus, we obtain:

$$p = \frac{2}{3} c_{10} \frac{(\lambda^3 - J)}{J^{5/3} \lambda} \tag{36}$$

and the uniaxial stress therefore is:

$$\mathbf{\Pi}_{11} = 2c_{10} \frac{(\lambda^3 - J)}{J^{2/3} \lambda^2} \tag{37}$$

replacing p from Eqs. (36) into (35), we obtain a nonlinear equation to compute J by knowing λ. In Fig. 2, we plot the lateral stretch upon the tension stretch for the two functions g_1 and g_2. For this example, we take $c_{10} = 1$ and $k = 5/3$ MPa. Depending on the value of the parameters a, b and c, we observe an unphysical response in compression, as previously mentioned (a non-monotonic behavior in compression, cf. solid lines in Fig. 2) or a compressible behavior that is limited with limit values (dotted lines in Fig. 2). We can also notice that when the compressibility limit is reached, curves have the same slope as an incompressible response.

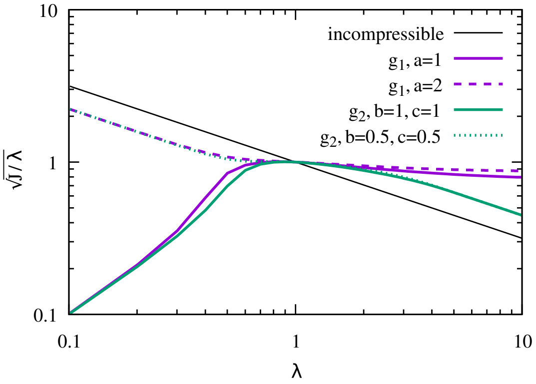

Fig. 2 Lateral stretch versus axial stretch for a simple compressible extension ($k/c_{10} = 5/3$)

4 Isothermal volumetric viscosity

We consider the trial case of an isothermal and adiabatic behavior with an isochoric elastic response and a dissipation in volume.

We propose a very simple model to take into account a volumetric viscosity with a hybrid energy approach. Starting from the intrinsic dissipation and using the previous isochoric/volumetric split, we assume that the term contracted with $\dot{\mathbf{F}}$ in Eq. (16) is null. Therefore:

$$\mathbf{\Pi} = \bar{\mathbf{\Pi}} : \mathbb{P}_{\mathbf{F}} - Jqh'(J)\mathbf{F}^{-T} \tag{38}$$

$$\phi^{\text{int}} = \underbrace{\left(h(J) + g'(q)\right)}_{A_q} \dot{q} \geq 0 \tag{39}$$

We now suppose that $A_q \neq 0$; therefore, we admit that the compressibility law is not fulfilled exactly at any time and that we have $h(J) \propto -g'(q)$ and therefore $\dot{J} \propto \dot{q}$ for a nearly incompressible material. We propose to consider the following complementary equation:

$$\boxed{A_q = \frac{\eta}{k}\dot{J}} \tag{40}$$

where η is a viscosity like parameter (dimension of a stress per time) such that $\eta << k$. As previously, we choose $h(J) = 1 - J$. Differentiating Eq. (40) with respect to time, we obtain the following:

$$-\dot{J} + g''(q)\dot{q} = \frac{\eta}{k}\ddot{J} \tag{41}$$

As $\eta << k$, we can neglect the right-hand side of Eq. (41) and we have $\dot{J} \simeq g''(q)\dot{q}$. Due to the convexity of $g(q)$, we recover the physical requirement of a compressibility/pressure relation: $\dot{J} \propto \dot{q}$. This shows that complementary Eq. (40) can be thermodynamically admissible (if $\eta << k$):

$$A_q\dot{q} = \frac{\eta}{k}\dot{J}\dot{q} \simeq \frac{\eta g''(q)}{k}\dot{q}^2 \geq 0 \quad \forall \dot{q} \tag{42}$$

To illustrate this model, we consider the same simple extension example as the one described in the previous section. Using the same isochoric hybrid energy and using the condition of uniaxiality of stress ($\mathbf{\Pi}_{22} = \mathbf{\Pi}_{33} = 0$), we obtain the following differential algebraic system to solve:

$$\begin{cases} \dfrac{\eta}{k}\dot{J} = 1 - J + g'(q) \\[2ex] q = \dfrac{2}{3}c_{10}\dfrac{\lambda^3 - J}{J^{5/3}\lambda} \end{cases} \tag{43}$$
$$q(t=0) = 0 \quad J(t=0) = 1$$

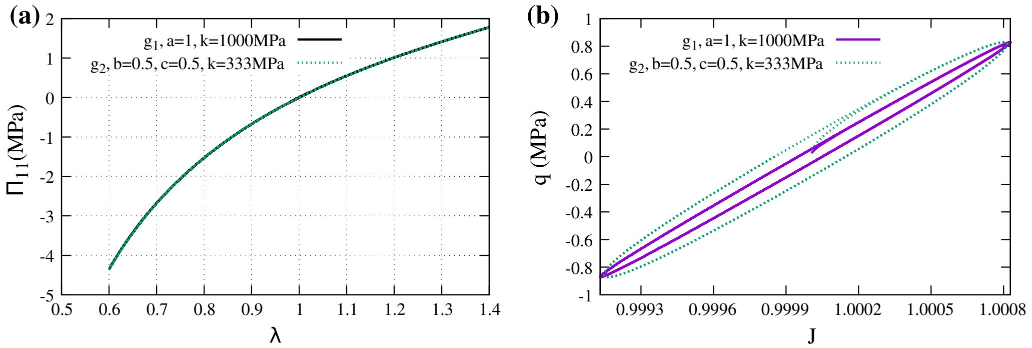

Fig. 3 Simple extension test with a volumetric viscosity (the modulus k is chosen such as to obtain the same infinitesimal bulk modulus value in each model.) **a** Uniaxial stress upon axial stretch. **b** Hydrostatic pressure upon volume variation

this system can be rewritten into the following shorter one:

$$\begin{aligned} &\frac{\eta}{k}\dot{J} = 1 - J + \hat{g}'(\lambda, J) \\ &J(t = 0) = 1 \end{aligned} \tag{44}$$

We consider a cyclic sinusoidal loading, $\lambda(t) = 1 + 0.4\sin(2\pi t)$, with $c_{10} = 1\,\mathrm{MPa}$, $\eta = 10\,\mathrm{MPa}\cdot s$. The results are shown in Fig. 3. The volumetric behavior exhibits a hysteresis loop that depends on the function g. We have adapted the value of the compressibility modulus for each model so as to have the same infinitesimal compressibility modulus. We can notice that the uniaxial stress/strain behavior remains elastic. This simple example illustrates that the hybrid energy approach allows a straightforward definition of dissipative volumetric behavior without introducing an additional variable.

This approach could be used within a standard modeling of isochoric viscosity such as for generalized Maxwell models or within other irreversible isochoric mechanisms. It could be applied to porous rubber materials, for instance, to model both the viscosity in volume and in isochoric modes such as done in [14] with separate internal variables.

5 Entropic elasticity

5.1 Heat balance

In this section, we consider a non-isothermal state. We assume that the transformation can be split into a pure thermal expansion and a pure mechanical deformation. The intermediate thermal state free of mechanical deformation is assumed to be stress free. This stress-free hypothesis is fundamental and motivates the following multiplicative decomposition:

$$\mathbf{F} = \mathbf{F}_\Theta \mathbf{F}_m \tag{45}$$

where $\mathbf{F}_\Theta = J_\Theta^{1/3}\mathbf{1}$ is a pure thermal expansion and $\mathbf{F}_m$ is the mechanical deformation gradient (the same approach has been adopted by many authors, e.g., see [12]). Using the isochoric-volumetric split of the total deformation gradient $\mathbf{F} = (J^{1/3}\mathbf{1})\bar{\mathbf{F}}$, it can be seen that:

$$J = J_\Theta J_m \quad \text{and} \quad \mathbf{F}_m = \bar{\mathbf{F}} \tag{46}$$

The separation of the deformation into a mechanical and a thermal contribution can be viewed as a generalization of the Duhamel–Neumann hypothesis for a nonlinear theory.

A very common extension of this idea of decomposition of thermal and mechanical contributions is to suppose an additive decomposition of the hybrid energy into a purely thermal part and a mechanical part (which could depend on temperature). As previously, and for the same reason (orders of magnitude of the strain energy), the mechanical part is assumed to be split into an isochoric and a volumetric contribution:

$$\varphi(\bar{\mathbf{F}}, q, \Theta) = \varphi_{\mathrm{iso}}(\bar{\mathbf{F}}, \Theta) + \varphi_{\mathrm{vol}}(q, \Theta) + \varphi_{\mathrm{ther}}(\Theta) = \varphi_{\mathrm{iso}}(\bar{\mathbf{F}}, \Theta) - \frac{g(q, \Theta)}{\rho_0} + \varphi_{\mathrm{ther}}(\Theta) \tag{47}$$

For the potential β, we assume that the stress variable q is directly related to the mechanical volumetric variation J_m. We thus propose to consider the choice:

$$\beta(J, q, \Theta) = q\frac{h(J_m)}{\rho_0} \tag{48}$$

Functions g and h can be chosen as in the previous sections. In the following, we will assume that g does not depend on temperature, but it can be considered that, for instance, the bulk modulus is temperature dependent. Using Eq. (13), and assuming $h(J_m) = 1 - J_m$ we obtain the following expression for the entropy:

$$\boxed{s = -\frac{\partial\varphi_{\text{iso}}}{\partial\Theta} - \frac{\partial\varphi_{\text{ther}}}{\partial\Theta} + \frac{1}{\rho_0}\frac{\partial g}{\partial\Theta} + \frac{Jq}{J_\Theta^2\rho_0}\frac{\partial J_\Theta}{\partial\Theta}} \tag{49}$$

Assuming an elastic behavior, i.e., $\phi^{\text{int}} = 0$, we obtain from Eq. (14):

$$\boxed{\begin{aligned} \mathbf{\Pi} &= \rho_0\frac{\partial\varphi_{\text{iso}}}{\partial\bar{\mathbf{F}}} : \mathbb{P}_{\mathbf{F}} + \frac{J}{J_\Theta}q\mathbf{F}^{-\mathrm{T}} && \text{(50a)}\\ \frac{\partial g}{\partial q} &= \left(\frac{J}{J_\Theta} - 1\right) = J_m - 1 && \text{(50b)} \end{aligned}}$$

In Eq. (50a), we can notice that the hydrostatic pressure p is related to q through: $p = q/J_\Theta$. Equation (50b) can be viewed as a compressibility law formulated with the mechanical volume variation rather than the total volume variation. Furthermore, if one adopts the same functions g as previously defined, it can be seen that a pure thermal expansion for which $\mathbf{F} = J_\Theta^{1/3}\mathbf{1}$ leads to $J_m = 1$ and $\bar{\mathbf{F}} = \mathbf{1}$; therefore, Eqs. (50a) and (50b) give the expected stress-free solution: $\mathbf{\Pi} = \mathbf{0}$, $q = 0$. This stress-free solution is obtained without any modification of the free energy, which would not be the case if, for instance, one chooses a potential β such that: $\beta = q(1 - J)$.

Combining Eqs. (6) and (4), we obtain:

$$\rho_0\dot{s}\Theta = \mathbf{\Pi} : \dot{\mathbf{F}} + \rho_0 r - \operatorname{div}_X\mathbf{Q}_\Theta - \rho_0\dot{\varphi} + \rho_0\dot{\beta} - \rho_0 s\dot{\Theta} \tag{51}$$

Using Eqs. (8) and (9) and regrouping terms, we have:

$$\rho_0\dot{s}\Theta = \phi^{\text{int}} + \rho_0 r - \operatorname{div}_X\mathbf{Q}_\Theta \tag{52}$$

The variation of the entropy can be computed from (49):

$$\begin{aligned} \dot{s} = &-\left(\frac{\partial^2\varphi_{\text{ther}}}{\partial\Theta^2} + \frac{\partial^2\varphi_{\text{iso}}}{\partial\Theta^2} - \frac{1}{\rho_0}\frac{\partial^2 g}{\partial\Theta^2} + \frac{2Jq}{J_\Theta^3\rho_0}\left(\frac{\partial J_\Theta}{\partial\Theta}\right)^2 - \frac{Jq}{J_\Theta^2\rho_0}\frac{\partial^2 J_\Theta}{\partial\Theta^2}\right)\dot{\Theta}\\ &-\left(\frac{\partial^2\varphi_{\text{iso}}}{\partial\Theta\partial\bar{\mathbf{F}}} : \mathbb{P}_{\mathbf{F}} - \frac{Jq\mathbf{F}^{-\mathrm{T}}}{J_\Theta^2\rho_0}\frac{\partial J_\Theta}{\partial\Theta}\right) : \dot{\mathbf{F}} + \frac{1}{\rho_0}\left(\frac{\partial^2 g}{\partial\Theta\partial q} + \frac{J}{J_\Theta^2}\frac{\partial J_\Theta}{\partial\Theta}\right)\dot{q} \end{aligned} \tag{53}$$

putting (53) in (52) and recalling that $\phi^{\text{int}} = 0$ in the thermoelastic case, we obtain the heat transfer equation:

$$\boxed{\rho_0 C_p\dot{\Theta} = l_m + l_q + \rho_0 r - \operatorname{div}_X\mathbf{Q}_\Theta} \tag{54}$$

where C_p is the isobaric heat capacity

$$C_p = -\Theta\left(\frac{\partial^2\varphi_{\text{ther}}}{\partial\Theta^2} + \frac{\partial^2\varphi_{\text{iso}}}{\partial\Theta^2} - \frac{1}{\rho_0}\frac{\partial^2 g}{\partial\Theta^2} + \frac{2Jq}{J_\Theta^3\rho_0}\left(\frac{\partial J_\Theta}{\partial\Theta}\right)^2 - \frac{Jq}{J_\Theta^2\rho_0}\frac{\partial^2 J_\Theta}{\partial\Theta^2}\right) \tag{55}$$

and l_m, l_q are mechanical coupling terms

$$\begin{aligned} l_m &= \Theta\left(\rho_0\frac{\partial^2\varphi_{\text{iso}}}{\partial\Theta\partial\bar{\mathbf{F}}} : \mathbb{P}_{\mathbf{F}} - \frac{Jq\mathbf{F}^{-\mathrm{T}}}{J_\Theta^2}\frac{\partial J_\Theta}{\partial\Theta}\right) : \dot{\mathbf{F}} = \Theta\frac{\partial\mathbf{\Pi}}{\partial\Theta} : \dot{\mathbf{F}}\\ l_q &= -\Theta\left(\frac{\partial^2 g}{\partial\Theta\partial q} + \frac{J}{J_\Theta^2}\frac{\partial J_\Theta}{\partial\Theta}\right)\dot{q} \end{aligned} \tag{56}$$

5.2 A simple thermoelastic energy (for rubberlike materials)

As an application, we consider the following isochoric and thermal potentials:

$$\rho_0\varphi_{\text{iso}} = c_{10}\frac{\Theta}{\Theta_0}(\bar{I}_1 - 3), \quad \rho_0\varphi_{\text{ther}} = C_0\left(\Theta - \Theta_0 - \Theta\log\left(\frac{\Theta}{\Theta_0}\right)\right) - C_1\frac{(\Theta - \Theta_0)^2}{2\Theta_0} \tag{57}$$

where c_{10}, C_0 and C_1 are material coefficients. For the thermal volume variation, we consider a very simple linear expansion relation:

$$J_\Theta = 1 + \alpha(\Theta - \Theta_0) \tag{58}$$

where α is the volumetric expansion coefficient and Θ_0 is the initial temperature. The proposed structure of the hybrid free energy obtained from Eqs. (47) and (57) can be viewed as an extension of the modified entropic theory earlier proposed by [4] (see [3] for the strictly entropic theory and [12,20] for the modified entropic theory within volumetric/isochoric splitting). The main motivation of it is that, first, the isochoric behavior is mainly entropic in rubberlike materials, and second, we want to be able to describe free-stress expansion of the media under a thermal variation. Therefore, we need an energetic volumetric contribution such as proposed in the modified entropic theory. To the opposite of the proposition of [4], we do not assume an additive decomposition of the internal volumetric energy in pure mechanical and thermal contributions. However, this additive decomposition can be recovered if one consider small temperature variations (or small expansion coefficient). In this case, J_m can be linearized according to: $J_m = J + \alpha(\Theta - \Theta_0) + o(\alpha(\Theta - \Theta_0))$.

Assuming that the function g does not depend on temperature [we take g as in Eqs. (24)–(25)], the isobaric heat capacity takes the following form:

$$C_p = \frac{1}{\rho_0}\left(C_0 + C_1\frac{\Theta}{\Theta_0} - \frac{2\alpha^2 Jq\Theta}{J_\Theta^3}\right) = \frac{1}{\rho_0}\left(C_0 + C_1\frac{\Theta}{\Theta_0} - \frac{2\alpha^2 Jp\Theta}{J_\Theta^2}\right) \tag{59}$$

From expression (59), we can see that the isobaric heat capacity can become negative if the term $2\alpha^2 p\Theta$ becomes larger than the linear term in temperature. This only occurs under strong hydrostatic tension and leads to a non-admissible heat capacity. However, in practice for rubber reaching this limit value will never be the case because it will lead to unrealistic value of hydrostatic tension: assuming that J and J_θ are closed to 1 and taking $\alpha = 6.10^{-4}\,\text{K}^{-1}$, $\Theta_0 = 293\,\text{K}$, $C_0 = 1 \times 10^6\,\text{J/m}^3 K$, $C_1 = 0.6 \times 10^6\,\text{J/m}^3\,\text{K}$, and $T = 380\,\text{K}$ we obtain a limit value of $q = 7\,\text{GPa}$ which clearly irrelevant for rubber.

With the previous assumptions, the coupling terms l_m, l_q take the following forms:

$$\begin{aligned} l_m &= \Theta\left(2\frac{c_{10}}{\Theta_0}J^{-2/3}(\mathbf{F} - \frac{I_1}{3}\mathbf{F}^{-\mathrm{T}}) - \frac{Jq\mathbf{F}^{-T}\alpha}{J_\Theta^2}\right) : \dot{\mathbf{F}} \\ l_q &= -\Theta\left(\alpha\frac{J}{J_\Theta^2}\right)\dot{q} \end{aligned} \tag{60}$$

5.3 Adiabatic stretching of a nearly incompressible rubber band (thermoelastic inversion)

We consider the standard example of homogeneous and adiabatic stretching of a rubber band similar to the one described in [12], but in the present case we make no hypothesis of perfect incompressibility of the material. We assume a uniaxial stretch λ and the deformation gradient is given by: $\mathbf{F} = \lambda\vec{e}_1 \otimes \vec{e}_1 + \sqrt{J/\lambda}\,\vec{e}_2 \otimes \vec{e}_2 + \sqrt{J/\lambda}\,\vec{e}_3 \otimes \vec{e}_3$, where λ and J depends on time. The stress and the hydrostatic pressure are given in this case by:

$$\mathbf{\Pi} = 2c_{10}\frac{\Theta}{\Theta_0}J^{-2/3}\left(\mathbf{F} - \frac{I_1}{3}\mathbf{F}^{-\mathrm{T}}\right) + q\frac{J}{J_\Theta}\mathbf{F}^{-\mathrm{T}} \tag{61}$$

$$\left(\frac{J}{J_\Theta} - 1\right) = g_1'(q) \quad \text{or} \quad \left(\frac{J}{J_\Theta} - 1\right) = g_2'(q) \tag{62}$$

Table 1 Material parameters

Density	ρ_0(Kg/m^3) 1000			
Thermal parameters	α(K^{-1}) $6 \cdot e^{-4}$	C_0(J/m^3/ K) $1.6e^6$	C_1(J/m^3/ K) $3.6e^3$	$\Theta_0(K)$ 293
Mechanical parameters	c_{10}(Pa) $0.22e^6$			

The uniaxial stress condition $\mathbf{\Pi}_{22} = \mathbf{\Pi}_{33} = 0$ gives a relation between Θ, J and q:

$$q = \frac{2}{3} J_\Theta \frac{c_{10}\Theta}{\Theta_0} \frac{\lambda^3 - J}{J^{5/3}\lambda} \tag{63}$$

The isobaric heat capacity is computed from Eq. (59):

$$\rho_0 C_p = C_0 + C_1 \frac{\Theta}{\Theta_0} + \frac{4c_{10}\alpha^2\Theta^2(J - \lambda^3)}{3J^{2/3}\Theta_0 \lambda J_\Theta^2} \tag{64}$$

and the coupling terms from Eqs. (60):

$$\begin{aligned} l_m &= 2c_{10}\Theta(J - \lambda^3)\frac{\lambda(\alpha\Theta + J_\Theta)\dot{J} - 3JJ_\Theta\dot{\lambda}}{3J^{5/3}\Theta_0\lambda^2 J_\Theta} \\ l_q &= -\Theta\left(\alpha\frac{J}{J_\Theta^2}\right)\left(\frac{2c_{10}\Theta(2J - 5\lambda^3)J_\Theta}{9J^{8/3}\Theta_0\lambda}\dot{J} - \frac{2c_{10}(J - \lambda^2)(\alpha\Theta + J_\Theta)}{3J^{5/3}\Theta_0\lambda}\dot{\Theta}\right. \\ &\quad \left. + \frac{2c_{10}\Theta(J + 2\lambda^3)J_\Theta}{3J^{5/3}\Theta_0\lambda^2}\dot{\lambda}\right) \end{aligned} \tag{65}$$

Assuming a perfect adiabatic case and considering an extension such that $\lambda(t) = 1 + \lambda_0\left(\frac{t}{T}\right)$ where $t \in [0, T]$, the equilibrium solution is found by solving the following system of two equations with two unknowns:

$$\text{found } J, \Theta \text{ such that:} \\ \begin{cases} \rho_0 C_p \dot{\Theta} = l_m + l_q \\ \dfrac{J}{J_\Theta} - 1 = g_1'(q) \quad \text{or} \quad \dfrac{J}{J_\Theta} - 1 = g_2'(q) \quad \text{with} \quad q = \dfrac{2}{3}J_\Theta\dfrac{c_{10}\Theta}{\Theta_0}\dfrac{\lambda^3 - J}{J^{5/3}\lambda} \end{cases} \tag{66}$$

We have solved the previous nonlinear differential algebraic equation system with Mathematica. The chosen material parameters are given in Table 1.

Figure 4 shows the temperature variation upon the extension for various values of the bulk modulus. We recover the so-called thermal elastic inversion phenomenon, which is due to a competition between the effect of the thermal expansion and the entropic nature of rubber elasticity. It can be noticed that the thermal inversion point is slightly dependent on the bulk modulus.

5.4 Adiabatic hydrostatic compression of a nearly incompressible rubber block

For this example, we consider the following deformation gradient: $\mathbf{F} = J^{1/3}\mathbf{I}$ and we impose a homogenous hydrostatic compression. Using Eqs. (50a) and (50b), we obtain:

$$\mathbf{\Pi} = \frac{J^{2/3}}{J_\Theta} q\mathbf{1} \tag{67}$$

$$\frac{J}{J_\Theta} - 1 = g'(q) \tag{68}$$

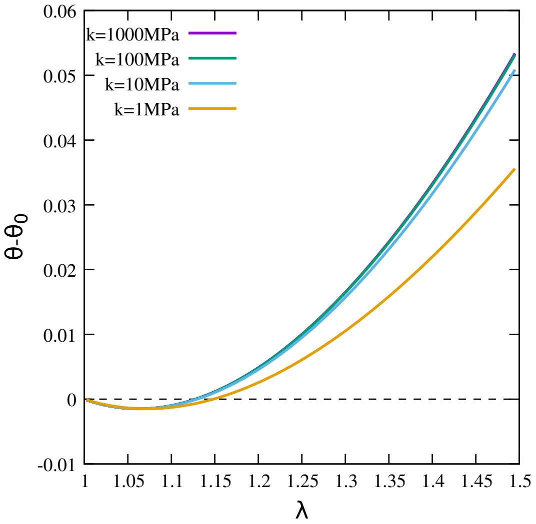

Fig. 4 Temperature variation for a simple extension: sensitivity to the bulk modulus for function $g_1(q)$ with $a = 1$

From Eq. (68), we can express J as a function of Θ and q. The isobaric heat capacity is given by:

$$\rho_0 C_p = C_0 + C_1 \frac{\Theta}{\Theta_0} + \frac{2\alpha^2 \Theta (1 + g'(q))q}{J_\Theta^2} \tag{69}$$

The coupling terms are expressed in this case as follows:

$$\begin{aligned} l_m &= -\frac{\alpha q \Theta}{J_\Theta^2}(J_\Theta g''(q)\dot{q} + \alpha(1 + g'(q))\dot{\Theta}) \\ lq &= -\frac{\alpha \Theta}{J_\Theta}(1 + g'(q))\dot{q} \end{aligned} \tag{70}$$

we consider the following pressure signal:

$$p(t) = \frac{q(t)}{J_\Theta(t)} = -p_0 t \text{ with } t \in [0, 1] \tag{71}$$

As for the previous example, we formulate the equilibrium solutions from the resolution of the following system of two equations with two unknowns:

$$\text{found } q, \Theta \text{ such that:} \\ \begin{cases} \rho_0 C_p \dot{\Theta} = l_m + l_q \\ \dfrac{\dot{q}}{J_\Theta} - \dfrac{q}{J_\Theta^2}\alpha\dot{\Theta} = p'(t) \end{cases} \tag{72}$$

We use the same numerical solver as in the previous example (function NDSolve of Mathematica). Figure 6 shows the results obtained for the two functions $g1$ and $g2$ and for different values of compressibility parameters. First, it can be noticed that the results are qualitatively in accordance with those obtained experimentally. In [23], the authors realized measurement of the temperature variation of a block of unvulcanized SBR under rapid hydrostatic pressure variation at different reference temperatures. They showed that the temperature variation depends nonlinearly upon the pressure variation as can be seen in Fig. 5.

Figure 6 illustrates the results obtained for the two compressibility functions $g_1(q)$ and $g_2(q)$ for various parameters of compressibility. It can be seen first that, depending on the parameter values, if the hydrostatic pressure is higher than the bulk modulus (which is irrelevent for standard rubber materials), we can obtain a non-monotonic behavior. When the hydrostatic pressure is lower than the bulk modulus, both functions $g_1(q)$ and $g_2(q)$ give results that are in accordance (same order of magnitude) with the experiment results. The hybrid free energy allows us to play with the nonlinearity in pressure in a more practical way than in the standard Helmholtz free energy case.

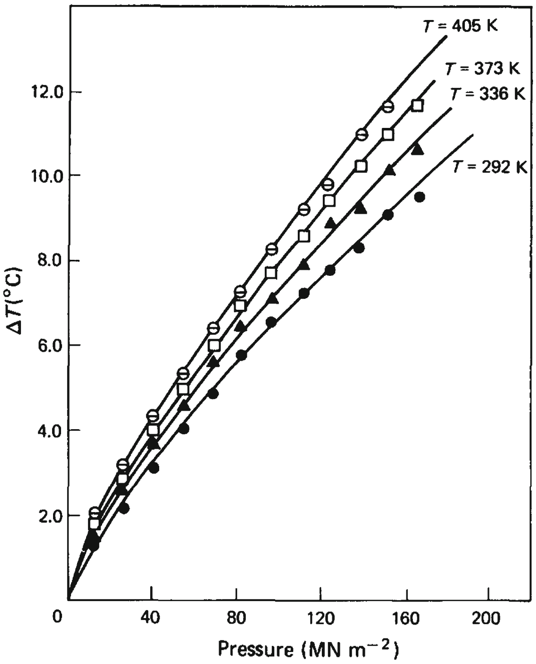

Fig. 5 Temperature changes as a function of applied pressure for styrene–butadiene rubber at different reference temperatures from [23]

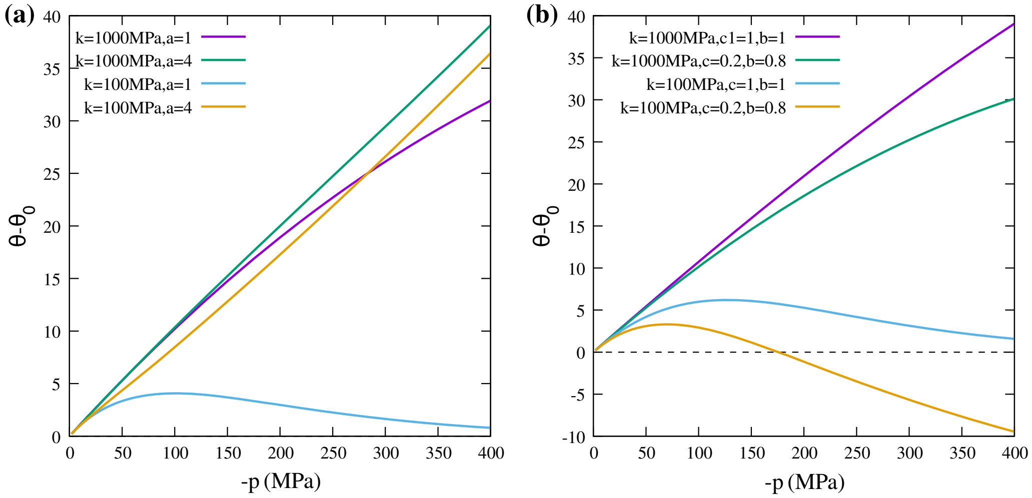

Fig. 6 Temperature variation for a hydrostatic and adiabatic compression. **a** Case $g(q) = g1(q)$. **b** Case $g(q) = g2(q)$

6 Conclusion

The hybrid or mixed free energy discussed in this work makes it possible to propose new volumetric energy functions defined from a pressure-like quantity rather than from a volume variation. It opens up new tracks for the development of constitutive models to address different issues. Standard nonlinear elasticity with nearly incompressible behavior or thermomechanical couplings can be taken into account with the advantage of obtaining a formulation where the pressure-like quantity is introduced from a thermodynamic potential. Thermoelastic couplings associated with volumetric deformations, for example, can be described and are in good qualitative agreement with the experimental results. Hybrid energy also makes it possible to obtain multi-field variational formulation directly without considering specific variational principles such as Hu–Washizu or Hellinger–Reissner. The pressure-like variable plays the role of a Lagrange multiplier, as in the previously mentioned principles without introducing a perturbation parameter. Volume dissipative mechanisms like volume viscosity can also be modelized without introducing further internal variables. As already shown in [16], a mixed free energy is also very attractive for the identification of the material parameters of thermomechanical models from constant pressure calorimetry experiments such as differential scanning calorimetry (DSC). Finally, this approach makes it possible to consider nearly incompressible behaviors from a new perspective.

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