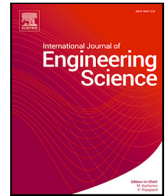


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Thermo-mechanically coupled constitutive equations for soft elastomers with arbitrary initial states

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ABSTRACT

It is a long-standing challenge to predict the thermo-mechanically coupled behaviors of initially stressed soft elastomers since most of the existing theories ignore the influences of thermoelastic deformation histories. The constitutive equations may be completely different even for the same initial stresses, if the latter is originated from isothermal and adiabatic deformations, respectively. In this paper, we establish a general framework for deriving constitutive equations for soft elastomers with arbitrary initial states. Instead of using the virtual stress-free configuration, we define the natural state by imposing the stress-free condition and the natural temperature condition. The derivations are based on a new proposed intrinsic embedding method of initial states, in which an additive decomposition of material strains is employed and the material coordinates can be properly defined. Once the natural-state-based free energy density and internal constraint are specified, the required constitutive equations can be accordingly obtained. We then derive the explicit formulations of the Cauchy stress and the entropy by linearization. On this basis, the embedding of initial states in Saint Venant–Kirchhoff, Blatz–Ko, Mooney–Rivlin, Neo-Hookean, Gent, and exponential form elastomers are detailed discussed. The influences brought by the initial stresses, the initial temperature, and the internal constraint on the elastic coefficients are analyzed separately. The new proposed constitutive equations show quantitative agreement with the classical theories under isothermal circumstances and fill a theoretical blank in this field under non-isothermal circumstances. Our approaches significantly improve the current constitutive theory of soft materials and may shed some light on the theoretical modeling of multi-field coupling problems.

1. Introduction

Soft materials including rubbers, foams, gels, living matter, granular matter, and organic matter, have received a wide range of applications. From traditional rubber-like products (Beatty, 1987) to modern soft robots (Martinez, Glavan, Keplinger, Oyetibo, & Whitesides, 2014), electronics and transducers (Bustamante, Shariff, & Hossain, 2021; Carpi et al., 2015; Kim, Ghaffari, Lu, & Rogers, 2012; Rogers, Someya, & Huang, 2010), biomedical modeling (Gao, Li, Wang, & Feng, 2021; Yin, Li, & Feng, 2021), and unconventional energies (Wang, Huang, Gao, & Zhao, 2021; Wang, Huang, Lin, & Zhao, 2019; Wang & Zhao, 2020). The first and foremost problem is to accurately describe the mechanical responses of these materials (Anssari-Benam, Bucchi, & Saccomandi, 2021; Boyce & Arruda, 2000; Destrade, Saccomandi, & Sgura, 2017; Marckmann & Verron, 2006; Signorini, 1961; Truesdell, 1956; Zhao, 2018). Undoubtedly, hyperelastic constitutive relations are the simplest and most prevalent ones, which requires only the

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specification of strain energies and internal constraints. To establish the corresponding theoretical formulations which agree with the experimental results, most of the existing constitutive models employ the stress-free hypothesis of reference configurations (Agoras, Lopez-Pamies, & Ponte Castañeda, 2009; Arruda & Boyce, 1993; Beatty, 2003; Bechir, Chevalier, Chaouche, & Boufala, 2006; Blatz & Ko, 1962; Dal, Açıkgöz, & Badienia, 2021; Gent, 1996; Hart-Smith, 1966; James, Green, & Simpson, 1975; Kachanov, 1992; Külcü, 2020; Lopez-pamies, 2010; Mooney, 1940; Ogden, 1972; Pucci & Saccomandi, 2002; Rajagopal & Saccomandi, 2016; Rivlin, 1948; Shariff, 2000; Treloar, 1943; Xiang et al., 2018; Yeoh, 1990). These models have become the mainstream of mechanical analysis of soft materials, for physical consistency and practicality.

Nonetheless, few materials undertake no initial stress (which reduces to residual stress when no initial traction is exerted). Leaving the most common gravity aside, initial stresses can be induced by non-uniform inelastic deformations, phase changes, surface modifications, heating or cooling, chemical reactions, and even geological movements (Chen & Eberth, 2012; Chuong & Fung, 1983; Cowin, 2006; Eskandari & Kuhl, 2015; Heibach et al., 2018; Hosford, 2010; Stylianopoulos et al., 2012; Withers & Bhadeshia, 2001b; Zang & Stephansson, 2010). These desirable or undesirable initial stresses have notable effects on the mechanical behaviors of soft materials, such as fatigue strength, fracture toughness, structure stability, crack morphology, wear, and corrosion (James, 2011; Masubuchi, 1980; Severson & Assadi, 2013; Shen & Zhao, 2018; Sun, Shen, & Zhao, 2019; Suo, Yang, & Shen, 2015; Totten, Howes, & Inoue, 2002; Wang, Zhao, & Huang, 2010; Webster & Ezeilo, 2001). They also serve some unique functions, for example, maintaining internal mechanical conditions, influencing the morphogenesis and growth rate of bio-tissues, and inducing buckling during evaporation (Ball, 2004; Ben-Amar & Goriely, 2005; Chuong & Fung, 1986; Fung, 1991; Holzapfel, Gasser, & Ogden, 2000; Li, Cao, Feng, & Gao, 2011; Merodio & Ogden, 2016; Nam, Park, & Ko, 2012; Taber, 1998; Taber & Humphrey, 2001; Wang et al., 2014). Because of this, the classical hyperelastic models are no longer appropriate. Proposing new constitutive relations that the influences of initial stresses are included becomes the center of attention.

The magnitude and distribution of initial stresses can be obtained from various destructive or non-destructive measurements (Brinksmeier et al., 1982; Greco, Sgambitterra, & Furgiuele, 2021; Hoger, 1985, 1986; Huang, Liu, & Xie, 2013; Rossini, Dassisti, Benyounis, & Olabi, 2012; Schajer, 2013; Withers & Bhadeshia, 2001a). Thus one only needs to embed the given initial stress σ_0 in the strain energy of soft elastomers. In this regard, there are three main embedding methods. The first approach is based on the theory of representations for tensor functions (Zheng, 1994), the strain energies therein are presupposed to be the functions of both strains and the given initial stresses, while the latter just act as additional variables (Agosti, Gower, & Ciarletta, 2018; Ciarletta, Destrade, & Gower, 2016; Ciarletta, Destrade, Gower, & Taffetani, 2016; Hoger, 1993a, 1993b; Joshi & Walton, 2013; Merodio, Ogden, & Rodriguez, 2013; Ogden & Singh, 2011; Shams, Destrade, & Ogden, 2011). The specific form of the strain energy is expressed as

$$W = W(F_e, \sigma_0), \quad (1.1)$$

where F_e is the subsequent deformation gradient. One can construct various combined invariants of F_e and σ_0 according to the behaved material symmetry (Spencer, 1971), and then formulate the corresponding strain energy. The advantage of this approach is that it requires no information about the origin of the given initial stresses, thus can be applied to complex situations.

The second approach is by considering the implicit constitutive theory proposed by Rajagopal and co-workers (Bustamante & Rajagopal, 2010, 2015a, 2015b, 2018; Rajagopal, 2003, 2007, 2010, 2011, 2015; Rajagopal & Saravanan, 2012; Rajagopal & Srinivasa, 2007, 2009). Different from the classical forms of constitutive equations of Cauchy elasticity, F_e here are given as the explicit functions of the Cauchy stress σ , but not the other way round, i.e.,

$$F_e = \mathfrak{H}(\sigma). \quad (1.2)$$

By this means, it is simpler and more convenient for embedding the given initial stresses, since the only additional condition that needs to be satisfied is that when the Cauchy stresses are equal to the initial stresses, the corresponding deformation gradients reduce to identity, namely, $\mathfrak{H}(\sigma_0) = I$. This is essentially an inverse problem in classical Cauchy elasticity.

It is noteworthy that the above two approaches require extra simulations and experiments to determine the presupposed material coefficients therein, thus, in general, the explicit forms of the functional expressions of constitutive relations remain unknown. To derive explicit constitutive equations that are convenient for direct applications, the third approach has appeared, which aims to model the whole process that produces the initial stresses (Chen & Hoger, 2000; Fung, 1990; Guillou & Ogden, 2006; Humphrey & Rajagopal, 2002; Lubarda & Hoger, 2002; Rajagopal & Srinivasa, 1998; Volokh, 2006). In particular, if one knows only the strain energies for initially stress-free hyperelastic materials, the given initial stresses can be properly embedded by employing the method of multiplicative decomposition of deformation gradients (Ambrosi et al., 2011; Balbi, Kuhl, & Ciarletta, 2015; Du & Lü, 2017; Du, Lü, Chen, & Destrade, 2018; Holzapfel, 2000; Lemate, Feuillard, Le Clézio, & Lethiecq, 2006; Menzel & Kuhl, 2012; Rodriguez, Hoger, & McCulloch, 1994; Skalak, Zargaryan, Jain, Netti, & Hoger, 1996; Wang, Zhang, & Chen, 2017). That is, according to

$$F_0 = F_0(\sigma_0), \quad W = W(F), \quad F = F_e F_0, \quad (1.3)$$

the initial-stresses-embedded strain energy can be derived as

$$W(F_e, \sigma_0) = W(F_e F_0(\sigma_0)), \quad (1.4)$$

where F, F_0 are the total and the initial deformation gradient, respectively. Although the initial stresses are formulated by the prescribed strain energy, it does not imply that the former is a result of prior elastic deformation, since the corresponding initial deformation gradient F_0 may not satisfy the compatibility condition (Hoger, 1997). The latter leads to the definition of *virtual*

stress-free configuration (Johnson & Hoger, 1993, 1995, 1998), which is the central concept in the whole theory. This approach is practical and concise but powerful enough to accurately predict the mechanical behaviors of soft elastomers with residual stresses.

However, when trying to further apply the multiplicative-decomposition-based approach to more general situations, some inherent shortcomings in the mathematical construction of the virtual stress-free configuration gradually are exposed. As Johnson and Hoger have indicated, it is essentially an infinite set of discrete points and need not be determined explicitly or achievable experimentally. Such a discontinuous concept can provide convenience for the constitutive derivation, but create a dilemma for further employing the continuum mechanics. Namely, if the elastomer is naturally inhomogeneous, i.e., $W(F, X_0)$, the strain energy will explicitly be dependent on the natural material coordinates X_0 , which cannot be determined by the approach. To be specific, suppose x, X are the current and the reference material coordinates, then from

$$\frac{\partial x}{\partial X_0} = F, \quad \frac{\partial x}{\partial X} = F_c, \quad \frac{\partial X}{\partial X_0} = F_0, \quad (1.5)$$

one finds that the following Pfaff equation

$$dX_0 - F_0^{-1}(X)dX = 0 \quad (1.6)$$

is non-integrable when F_0 is not compatible, thus the corresponding X_0 is often multi-valued. That is, the non-differentiability of the fields defined over the virtual stress-free configuration originates from the multi-value of the natural material coordinates. If the given initial stress is incompatible, differentiation cannot be done over the stress-free configuration. Therefore, not only those naturally inhomogeneous elastomers can not be considered, but also it is meaningless to take the material derivative of the discontinuous initial strains and densities. The latter limits the extension of the approach to nonlocal theories and strain gradient models.

More importantly, remarkable thermal effects can be observed accompanied by the deformation of soft materials (Chester & Anand, 2011; Witten, 1999), thus considering merely initial stresses may not determine the subsequent mechanical behaviors. For example, the same initial stresses can be originated from initially isothermal or adiabatic deformations. The former is essentially the above approach that has no variation of temperature, while the latter keeps the entropy unchanged and is likely to correspond to completely different constitutive equations. Therefore, thermal effects are indispensable in improving and perfecting the embedding method of given initial stresses.

In the present paper, we depart from the above three approaches by proposing a new *intrinsic embedding method of initial states*. Here, the adjective “intrinsic” indicates that our embedding method is formulated purely in material forms (Sedov, 1966; Synge & Chien, 1941), irrespective of the extrinsic properties brought by the Euclidean space where the deformation takes place. And the use of the term “initial states” but not the classical “initial stresses” mostly because the variation of temperature is taken into account in our approach, where the state variables are stresses and temperature (or equivalently, strains and entropy). Thus an additive decomposition of material strains is naturally obtained based on the variation of Riemannian metrics of the corresponding material manifold. By this means, the stress-free state is replaced by a continuum equipped with non-Euclidean metrics, namely, the *natural state*, where the given initial stresses and initial variation of temperature are removed. In other words, the natural state means that the elastomer is simultaneously in a stress-free state and at a prescribed natural temperature. Figuratively speaking, we stick all the discrete points of stress-free configuration together by introducing nonzero Riemannian curvatures to the material manifold (Fig. 1). The same idea has appeared in constructing the geometric field theory of defects (Bilby, Bullough, & Smith, 1955; Kleinert, 1989; Kröner, 1981; Nagahama & Teisseyre, 2008), where affine connections and the corresponding curvatures and torsions are introduced to eliminate the defects. Once the initial deformations satisfy the compatibility condition, then the natural state can be physically implemented into the Euclidean space, and thus it degenerates to the classical natural configuration. Only under such circumstances can coordinate transformations upgrade to deformation gradient using the linear structures of the Euclidean space, and then the multiplicative decomposition (1.3) can be valid. On this basis, if the variation of temperature is further neglected, i.e., considers only isothermal deformations, the new proposed additive-decomposition-based approach coincides with the above multiplicative-decomposition-based approach.

In the classical work of Saravanan (2008), the author used the terminologies *stress-free reference configuration* and *stressed reference configuration*. They correspond to the stress-free configuration (or the natural state) and the reference configuration in our paper, respectively. However, the concept of *stress-free reference configuration* is essentially different from our concept of stress-free configuration (or the natural state). According to the definition of Saravanan (2008), the former is accessed through a non-dissipative process from the stressed reference configuration. Thus, it is a continuum in Euclidean space. Our concept of stress-free configuration (or the natural state) is accessed through thermo-mechanically coupled unloading so that the initial stress and the initial temperature are removed for any individual material element. Due to the arbitrariness of the initial stress and the initial temperature distribution, the corresponding initial deformation that produces such an initial state may not be compatible. Therefore, the material elements can only exist as a pile of discrete points in classical theories. Using our framework, i.e., the mapping between Riemannian and Euclidean spaces, this pile of discrete points becomes a continuum in Riemannian space.

Note that differentiation is always allowed on the *stress-free reference configuration* since it is a continuum in Euclidean space. However, differentiation is allowed on the stress-free configuration only and only if the latter has become a continuum in Riemannian space. In other words, the differentiability of the fields defined over the stress-free configuration can only be established in our framework. The direct use of stressed reference configuration can remove the need for mapping between Riemannian and Euclidean spaces, and the corresponding theory has been elegantly established by Saravanan (2008). However, such representation for constitutive relations from a stressed reference configuration is irrespective of the origin of the initial stresses. The constitutive

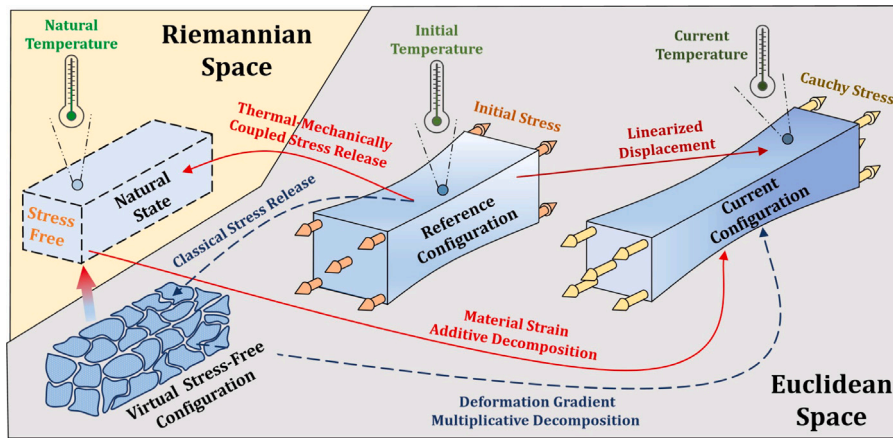


Fig. 1. Schematic diagram of the intrinsic embedding method of initial states.

equations may be completely different even for the same initial stress that experienced different thermo–mechanically coupled deformation histories. Therefore, our framework’s motivation is to take the origin of initial stress into account, where using Riemannian spaces is inevitable.

We then aim to formulate the thermo–mechanically coupled constitutive equations for soft elastomers with arbitrary initial states based on the newly proposed embedding method. Special attention is paid to the construction of the corresponding linearization theory, i.e., small displacements are superposed upon initially finite deformations. The center of this theory is to derive the associate elasticity tensor, which depends explicitly on the given initial states and is crucial in the study of wave propagation, structural dynamics, and elastic stability. Note that the linearization theory is different from the classical incremental theory, where some significant terms concerning the first order of displacement gradients may be missing. This is caused by the different orders of taking derivatives and carrying out linearization (Hoger & Johnson, 1995a, 1995b; Rajagopal, 2018; Tonon, 2002, 2010).

Our theory is inherently more complex than the abovementioned approaches because the intrinsic embedding method of initial states is based on rigorous mathematical derivations concerning Riemannian geometry. However, the new method has advantages in engineering practices for the following three reasons. The first reason is the full incorporation of thermal effects, which is crucial in modern engineering sciences and cannot be considered by the existing theories of initial stresses. In other words, the framework proposed in this paper fills a theoretical blank in this field under non-isothermal circumstances.

Second, our theory has established a unified framework for the constitutive modeling of initially stressed materials, not only thermoelastic elastomers. The embedding method of initial states is compatible with other developed continuum models because the natural state is a continuum in our theory but a pile of discrete points in the existing theories. For instance, it is convenient to establish the theory of residually stressed low-dimensional elastomers (rods, plates, shells, and membranes), initially stressed electromagnetic elastomers, liquid crystals with initial states, strain gradient elasticity with residual stresses, and others. Especially for the latter, gradients can only be performed in a continuum rather than a pile of discrete points. If we want to obtain explicit forms of thermo–mechanically coupled constitutive relations, the information about the origin of the residual stress is necessary, so we must carry out unloading and cannot directly use the stressed reference configuration. Thus, the need for mapping between Riemannian and Euclidean spaces cannot be removed.

Last but not least, this paper actually provides a modularized approach that is convenient to apply. In the existing theories that are based on the multiplicative decomposition of deformation gradients, the corresponding constitutive functions depend on the initial deformation gradient. Therefore, using these theories, the first thing is to express the initial deformation gradient by the given initial stress. The derivations of such expressions are often complicated and need to be carried out case by case since the existing theories are entirely based on matrix multiplication. Meanwhile, our additive decomposition-based framework is rather convenient. Although the constitutive functions depend on the natural material metrics, much effort has been made to carry out calculations and derive the algebraic and differential relationships between variables. Therefore, for engineers to use our theory, only two steps need to be followed: Firstly, to calculate the suggested coefficients and then construct the corresponding algebraic equations. The desired constitutive relations can be obtained once these equations are solved. In this respect, the present formulation has practical advantages.

The paper is arranged as follows. Section 2 outlines the preliminaries for the subsequent discussions, including the related deformation geometry and the forms of free energy density and internal constraint. In Section 3, we propose the intrinsic embedding method of initial states, which can be subdivided into two steps: first to establish the framework of additive decomposition of material strains and then use it to embed the given initial states to the free energy density and the internal constraints. Next, we derive explicitly in Section 4 the constitutive equations of the most common naturally isotropic elastomers. In Section 5, we linearize the obtained equations and present the general formulations of the Cauchy stresses and the entropy. Five illustrative examples including six prevailing hyperelastic models of Saint Venant–Kirchhoff, Blatz–Ko, Mooney–Rivlin, Neo-Hookean, Gent, and exponential form are detailed analyzed in Section 6. Also, the comparison between our new formulations and the multiplicative-decomposition-based and the implicit-constitutive-theory-based ones is carried out. Conclusions are provided in Section 7.

2. Preliminaries

2.1. Deformation geometry

Suppose B_0 is the given reference configuration, while B is the undetermined current configuration. They are both equipped with the same material coordinates x^i . The associated reference and current coordinate bases are e_i and ϵ_i , respectively. Thus reference material metrics can be derived as $g_{ij} = e_i \cdot e_j$, which have contravariant g^{ij} . Here, ϵ_{ij} denote strains while $G_{ij} = \epsilon_i \cdot \epsilon_j$ denotes current material metrics. Thus $G_{ij} = g_{ij} + 2\epsilon_{ij}$ and they have contravariant¹

$$\begin{aligned} G^{ij} &= \frac{1}{2G} \epsilon^{ikm} \epsilon^{j\ell q} G_{k\ell} G_{mq} = \frac{1}{2G} \epsilon^{ikm} \epsilon^{j\ell q} (g_{k\ell} + 2\epsilon_{k\ell}) (g_{mq} + 2\epsilon_{mq}) \\ &= \frac{(1 + 2I_1 + 4I_2) g^{ij} - 2(1 + 2I_1) I_1^{ij} + 4I_2^{ij}}{1 + 2I_1 + 4I_2 + 8I_3}, \end{aligned} \tag{2.1}$$

where ϵ^{ijk} is the 3D Levi-Civita symbol, the contravariant of g_{ij} is $g^{ij} = \frac{1}{2g} \epsilon^{ikm} \epsilon^{j\ell q} g_{k\ell} g_{mq}$, inversely, $g_{ij} = \frac{g}{2} \epsilon_{ikm} \epsilon_{j\ell q} g^{k\ell} g^{mq}$, and $g = \frac{1}{6} \epsilon^{ikm} \epsilon^{j\ell q} g_{ij} g_{k\ell} g_{mq}$ is the determinant of g_{ij} ,² I_1^{ij}, I_2^{ij} are polynomials of ϵ_{ij} of degree one and two

$$I_1^{ij} = g^{ik} g^{j\ell} \epsilon_{k\ell}, \quad I_2^{ij} = g^{ik} g^{j\ell} g^{mq} \epsilon_{km} \epsilon_{q\ell}, \tag{2.2}$$

while I_1, I_2, I_3 are three principal strain invariants

$$I_1 = g_{ij} I_1^{ij}, \quad I_2 = \frac{1}{2} (I_1^2 - \epsilon_{ij} I_1^{ij}), \quad I_3 = \frac{1}{3} (3I_1 I_2 - I_1^3 + \epsilon_{ij} I_2^{ij}). \tag{2.3}$$

and G is the determinant of G_{ij} , it can be derived as

$$\begin{aligned} G &= \frac{1}{6} \epsilon^{ikm} \epsilon^{j\ell q} G_{ij} G_{k\ell} G_{mq} \\ &= \frac{1}{6} \epsilon^{ikm} \epsilon^{j\ell q} (g_{ij} + 2\epsilon_{ij}) (g_{k\ell} + 2\epsilon_{k\ell}) (g_{mq} + 2\epsilon_{mq}) \\ &= g (1 + 2I_1 + 4I_2 + 8I_3). \end{aligned} \tag{2.4}$$

Given ρ_0 the reference mass density, then current mass densities ρ can be determined by continuity equation

$$\rho = \frac{\sqrt{g}}{\sqrt{G}} \rho_0 = \frac{\rho_0}{\sqrt{1 + 2I_1 + 4I_2 + 8I_3}}. \tag{2.5}$$

Suppose $u = u^i e_i$ is the displacement vector, then the transformations between the two different coordinate bases are specified in the form of

$$\epsilon_i = \left(\delta_i^j + \nabla_i u^j \right) e_j, \tag{2.6}$$

where δ_i^j are Kronecker symbols, and $\nabla_i u^j$ are displacement gradients, i.e.,

$$\nabla_i u^j = \frac{\partial u^j}{\partial x^i} + \Gamma_{ik}^j u^k, \quad \Gamma_{ik}^j = \frac{1}{2} g^{j\ell} \left(\frac{\partial g_{i\ell}}{\partial x^k} + \frac{\partial g_{k\ell}}{\partial x^i} - \frac{\partial g_{ik}}{\partial x^\ell} \right), \tag{2.7}$$

in which Γ_{ik}^j are Christoffel symbols. According to (2.6), strains ϵ_{ij} become

$$\epsilon_{ij} = \frac{1}{2} (G_{ij} - g_{ij}) = \frac{1}{2} (\nabla_i u_j + \nabla_j u_i + \nabla_i u_k \nabla_j u^k). \tag{2.8}$$

If there is no deformation, i.e., $\epsilon_{ij} = 0$, current quantities G_{ij}, G^{ij} and G will degenerate to g_{ij}, g^{ij} and g , respectively, while $\epsilon_i \neq e_i$ due to rigid rotations.

2.2. The free energy density and the internal constraint

The thermodynamic properties of the elastomer are specified by providing the following free energy density (per unit mass) and internal constraint

$$F = F(g_{ij}, \epsilon_{ij}, T, x^i), \quad C(g_{ij}, \epsilon_{ij}, T, x^i) = 0, \tag{2.9}$$

where T is the absolute temperature. If the elastomer is isotropic, (2.9) reduce to the following forms

$$F = F(I_1, I_2, I_3, T, x^i), \quad C(I_1, I_2, I_3, T, x^i) = 0. \tag{2.10}$$

¹ All the expressions and definitions presented in this section are the same as those in Sedov (1966). The convention of summation of Latin alphabet indices i, j, k, ℓ, m, q taken from 1 to 3 is adopted here. Note that coordinates x^i are often curvilinear on B even though they may be initially Cartesian on B_0 , thus it is necessary to distinguish covariant and contravariant tensors (equivalently, components with subscripts and superscripts).

² We here use an important identical equation of the product of two Levi-Civita symbols: $\epsilon^{ikm} \epsilon^{j\ell q} = g (g^{ij} g^{k\ell} g^{mq} + g^{i\ell} g^{kq} g^{mj} + g^{iq} g^{kj} g^{m\ell} - g^{iq} g^{k\ell} g^{mj} - g^{i\ell} g^{kj} g^{mq} - g^{ij} g^{kq} g^{m\ell})$.

According to Sedov (1966), the explicit dependency of g_{ij} in (2.9) is for achieving tensor contractions. To explain this in more detail, we consider (2.10), for instance, where I_1, I_2, I_3 are variables for \mathcal{F} and C and they are also the functions of g_{ij} and ε_{ij} (recall (2.2) and (2.3)). In this way, the material metrics g_{ij} , of course, become variables for \mathcal{F} and C . In general, material metrics are the core of Riemannian geometry and are crucial in contracting tensor components into scalar functions, especially in some hyper-elastic models (see (4.1) and (4.2), (6.25) and (6.26) below), different material metrics G_{ij}, g_{ij}^* will be used, not only g_{ij} . Thus, it is necessary to specify which material metrics will be used in a specific constitutive model. These reasons explain why we need to add g_{ij} as variables for \mathcal{F} and C .

The Cauchy stress tensor is expressed as

$$\sigma = \delta^{ij} \varepsilon_i \otimes \varepsilon_j = \delta^{k\ell} (\delta_k^i + \nabla_k u^i) (\delta_\ell^j + \nabla_\ell u^j) e_i \otimes e_j = \sigma^{ij} e_i \otimes e_j, \tag{2.11}$$

while the constraint multiplier and the mass entropy are denoted by p and s , respectively. Then the thermo-mechanically coupled constitutive equations of the elastomer can be derived in the following forms

$$\delta^{ij} = \rho \left(\frac{\partial \mathcal{F}}{\partial \varepsilon_{ij}} - \frac{p}{\rho} \frac{\partial C}{\partial \varepsilon_{ij}} \right), \quad s = - \left(\frac{\partial \mathcal{F}}{\partial T} - \frac{p}{\rho} \frac{\partial C}{\partial T} \right). \tag{2.12}$$

3. The intrinsic embedding method of initial states

3.1. The additive decomposition of material strains

There are two conditions to achieve the natural state B^* : the stress-free (mechanical) condition and the natural temperature (thermal) condition. To be specific, if reference configuration B_0 is not a natural state, there must exist initial stress $\sigma_0 = \sigma_0^{ij} e_i \otimes e_j$ or initial increment of temperature $\vartheta^* = T_0 - T^*$, where T_0 is the reference temperature of B_0 , while T^* is the given natural temperature of B^* . Namely, $B_0 = B^*$ only and only if $\sigma_0^{ij} = 0$ and $\vartheta^* = 0$.

If $B_0 \neq B^*$, free energy density and internal constraint (2.9) are no longer valid since they are made to describe the deformations that start from B^* , but not from B_0 . Thus it requires a modification so that σ_0^{ij} and ϑ^* can be embedded into (2.9) properly. By introducing the material metrics of B^* , i.e., g_{ij}^* , the material strains that start from B^* to B_0 and B_t can be derived as $\varepsilon_{ij}^* = \frac{1}{2}(g_{ij} - g_{ij}^*)$ and $\varepsilon_{ij}^{\text{tot}} = \frac{1}{2}(G_{ij} - g_{ij}^*)$, respectively. By this means, one obtains the following additive decomposition

$$\varepsilon_{ij}^{\text{tot}} = \varepsilon_{ij}^* + \varepsilon_{ij}. \tag{3.1}$$

The additive decomposition of material strains was first proposed by Sedov (1966). The original purpose of introducing such a decomposition is to distinguish elastic and plastic deformations in non-linear elastoplastic analysis. The subsequent works extended the additive decomposition to different types of material: elastoplasticity (Simo & Kennedy, 1992); thermoelasticity (Reddy & Chin, 1998); viscoelasticity (Lubarda, 2011); growth (Liang & Mahadevan, 2011); swelling (van der San, 2015), and surface defects (Roychowdhury & Gupta, 2018). It is accustomed to regard that only multiplicative decomposition can be applied to study finite deformations (Reina & Conti, 2014). However, no matter ε_{ij} or ε_{ij}^* are not assumed to be small in (3.1). The discord between the two types of decomposition methods is originated in the employing of distinct coordinates. Multiplicative decomposition (1.5) requires at least three different coordinates, i.e., X_0, X and x , correspond to B^*, B_0 and B , respectively. Only in this way can chain rules be applied properly and thus (1.3) be correct. However, different vector bases are associated with different coordinates, thus the strain components under different bases cannot be added together directly. That is why the classical additive decomposition can be regarded as approximately valid only when the differences between these bases are small enough. In our approach, there is only one type of coordinates, i.e., x^i , thus all the components of the material metric can be added directly without any additional requirements.

3.2. The embedding of initial states

According to (2.9) and (3.1), the free energy density and the internal constraint now can be modified to the following forms

$$\mathcal{F} = \mathcal{F}(g_{ij}^*, \varepsilon_{ij}^{\text{tot}}, T, x^i), \quad C(g_{ij}^*, \varepsilon_{ij}^{\text{tot}}, T, x^i) = 0. \tag{3.2}$$

Then from (2.12), the corresponding constitutive equations are expressed as³

$$\begin{aligned} \delta^{ij} &= \rho \left(\frac{\partial \mathcal{F}}{\partial \varepsilon_{ij}} - \frac{p}{\rho} \frac{\partial C}{\partial \varepsilon_{ij}} \right) (g_{ij} - 2\varepsilon_{ij}^*, \varepsilon_{ij}^* + \varepsilon_{ij}, T_0 + \vartheta, x^i), \\ s &= - \left(\frac{\partial \mathcal{F}}{\partial \vartheta} - \frac{p}{\rho} \frac{\partial C}{\partial \vartheta} \right) (g_{ij} - 2\varepsilon_{ij}^*, \varepsilon_{ij}^* + \varepsilon_{ij}, T_0 + \vartheta, x^i). \end{aligned} \tag{3.3}$$

Here, ρ_0, g_{ij}, T_0, T^* (or equivalently, ϑ^*) and σ_0^{ij} are given functions that act as initial conditions. Thus our main task is to express ε_{ij}^* in terms of these known quantities, i.e., to derive expressions $\varepsilon_{ij}^* = \varepsilon_{ij}^*(g_{ij}, \rho_0, \sigma_0^{ij}, \vartheta^*, T_0, x^i)$.

³ To avoid confusion, the terms in the brackets containing commas, for instance, $(*, *, *)$, are actually arguments of the expressions before them.

By substituting $\hat{\sigma}^{ij} = 0, \rho = \rho^*, p = p^*, \varepsilon_{ij}^{\text{tot}} = \vartheta = 0$ and $T_0 = T^*$ into (3.3), natural constraint multiplier $p^* = p^*(g_{ij}, \rho_0, \varepsilon_{ij}^*, T^*, x^i)$, natural entropy $s^* = s^*(g_{ij}, \rho_0, \varepsilon_{ij}^*, T^*, x^i)$ and natural mass density $\rho^* = \rho^*(g_{ij}, \rho_0, \varepsilon_{ij}^*, T^*, x^i)$ can be accordingly derived by solving the following algebraic equations

$$\begin{aligned} \left(\frac{\partial F}{\partial \varepsilon_{ij}} - \frac{p^*}{\rho^*} \frac{\partial C}{\partial \varepsilon_{ij}} \right) (g_{ij} - 2\varepsilon_{ij}^*, 0, T^*, x^i) &= 0, \\ \left(\frac{p^*}{\rho^*} \frac{\partial C}{\partial \vartheta} - \frac{\partial F}{\partial \vartheta} \right) (g_{ij} - 2\varepsilon_{ij}^*, 0, T^*, x^i) &= s^*, \\ \sqrt{g^* \rho^*} &= \sqrt{g} \rho_0. \end{aligned} \tag{3.4}$$

Similarly, by substituting $\hat{\sigma}^{ij} = \sigma_0^{ij}, \rho = \rho_0, p = p_0, \vartheta = 0$ and $\varepsilon_{ij} = 0$ into (3.3), one obtains the algebraic equations of ε_{ij}^* and p_0

$$\begin{aligned} \left(\frac{\partial F}{\partial \varepsilon_{ij}} - \frac{p_0}{\rho_0} \frac{\partial C}{\partial \varepsilon_{ij}} \right) (g_{ij} - 2\varepsilon_{ij}^*, \varepsilon_{ij}^*, T_0, x^i) &= \frac{\sigma_0^{ij}}{\rho_0}, \\ C(g_{ij} - 2\varepsilon_{ij}^*, \varepsilon_{ij}^*, T_0, x^i) &= 0. \end{aligned} \tag{3.5}$$

Therefore, according to distinct thermo-mechanically coupled deformation histories, simultaneous equations (3.4) and (3.5) can be inversely solved as

$$\varepsilon_{ij}^* = \varepsilon_{ij}^*(g_{ij}, \rho_0, \sigma_0^{ij}, \vartheta^*, T_0, x^i), \quad p_0 = p_0(g_{ij}, \rho_0, \sigma_0^{ij}, \vartheta^*, T_0, x^i). \tag{3.6}$$

Moreover, according to (3.3), initial entropy s_0 can be derived as

$$\begin{aligned} s_0 &= s_0(g_{ij}, \rho_0, \sigma_0^{ij}, \vartheta^*, T_0, x^i) \\ &= \left[\frac{\partial F}{\partial \vartheta} - \frac{p_0(g_{ij}, \rho_0, \sigma_0^{ij}, \vartheta^*, T_0, x^i)}{\rho_0} \frac{\partial C}{\partial \vartheta} \right] \Big|_{\varepsilon_{ij}^* = \varepsilon_{ij}^*(g_{ij}, \rho_0, \sigma_0^{ij}, \vartheta^*, T_0, x^i)}. \end{aligned} \tag{3.7}$$

Then we introduce the equivalent free energy density

$$\begin{aligned} \hat{F}(g_{ij}, \varepsilon_{ij}, \vartheta, \rho_0, \sigma_0^{ij}, \vartheta^*, T_0, x^i) \\ = F|_{\varepsilon_{ij}^* = \varepsilon_{ij}^*(g_{ij}, \rho_0, \sigma_0^{ij}, \vartheta^*, T_0, x^i)} + s_0(g_{ij}, \rho_0, \sigma_0^{ij}, \vartheta^*, T_0, x^i) \vartheta - \frac{\sigma_0^{ij} \varepsilon_{ij}}{\rho_0}, \end{aligned} \tag{3.8}$$

and the equivalent internal constraint

$$\hat{C}(g_{ij}, \varepsilon_{ij}, \vartheta, \rho_0, \sigma_0^{ij}, \vartheta^*, T_0, x^i) = C|_{\varepsilon_{ij}^* = \varepsilon_{ij}^*(g_{ij}, \rho_0, \sigma_0^{ij}, \vartheta^*, T_0, x^i)}, \tag{3.9}$$

where $\rho_0, \sigma_0^{ij}, \vartheta^*$ and T_0 serve as given parameters. By substituting (3.8) and (3.9) into (3.3), the required constitutive equations are expressed in the form of

$$\hat{\sigma}^{ij} = \frac{\rho}{\rho_0} \sigma_0^{ij} + \rho \left(\frac{\partial \hat{F}}{\partial \varepsilon_{ij}} - \frac{p}{\rho} \frac{\partial \hat{C}}{\partial \varepsilon_{ij}} \right), \quad s = s_0 - \left(\frac{\partial \hat{F}}{\partial \vartheta} - \frac{p}{\rho} \frac{\partial \hat{C}}{\partial \vartheta} \right). \tag{3.10}$$

Therefore, the problem of embedding the given initial states into (2.9) is converted to solve simultaneous algebraic equations (3.4) and (3.5). Also, the validity of the above embedding approach depends on the existence of the solutions of the equations. Whether the solutions exist or not is essentially a physical than a mathematical problem: if there is no solution for a certain type of elastomer, the contradiction should be attributed to the inadequate chosen of (2.9). Conversely, given certain forms of (2.9), they will restrict the possible distributions of initial states. It should be noted that the restrictions produced by the existence of the solutions are distinct from those by the compatibility of material strains. The latter is the major limitation of the further developments of the multiplicative-decomposition-based approach. However, in our new proposed additive-decomposition-based approach, material coordinates x^i are always existed no matter g_{ij}^* are Euclidean or not. In this respect, the intrinsic embedding method of initial states compares favorably with other methods.

4. The naturally isotropic elastomer

If the natural state of an elastomer is isotropic, it is a naturally isotropic elastomer. The presence of initial states often brings the anisotropy of the reference configuration (the initial-states-induced-anisotropy). When there is no initial stress or initial temperature increment, the reference configuration is the natural state, and the naturally isotropic elastomer is the isotropic elastomer in its usual sense. To naturally isotropic elastomers, (3.2) reduce to

$$F = F(I_1, I_2, I_3, T, x^i), \quad C = C(I_1, I_2, I_3, T, x^i), \tag{4.1}$$

where I_1, I_2, I_3 are three principal invariants of $\varepsilon_{ij}^{\text{tot}}$

$$\begin{aligned} I_1 &= g^{*ij} \varepsilon_{ij}^{\text{tot}}, \quad I_2 = \frac{1}{2} \left(I_1^2 - g^{*ik} g^{*jl} \varepsilon_{ij}^{\text{tot}} \varepsilon_{kl}^{\text{tot}} \right), \\ I_3 &= \frac{1}{3} \left(3I_1 I_2 - I_1^3 + g^{*ik} g^{*jl} g^{*rs} \varepsilon_{ij}^{\text{tot}} \varepsilon_{kr}^{\text{tot}} \varepsilon_{sl}^{\text{tot}} \right). \end{aligned} \tag{4.2}$$

Note that it is the contraction of $\varepsilon_{ij}^{\text{tot}}$ and g^{*ij} , but not g_{ij} , that constitutes the invariants. We here introduce the following contravariant strains

$$\mathcal{P}^{ij} = g^{*ij} - g^{ij} \tag{4.3}$$

to replace ε_{ij}^* . The reason why \mathcal{P}^{ij} is more convenient than ε_{ij}^* is that \mathcal{P}^{ij} and initial stresses σ_0^{ij} are both contravariant tensors. By substituting $g_{ij}^* = g_{ij} - 2\varepsilon_{ij}^*$ into (4.3), one obtains

$$\varepsilon_{ij}^* = \frac{\mathcal{P}_3 g_{ij} + g_{ik} g_{j\ell} [(1 + \mathcal{P}_1) \mathcal{P}^{k\ell} - Q^{k\ell}]}{2(1 + \mathcal{P}_1 + \mathcal{P}_2 + \mathcal{P}_3)}, \tag{4.4}$$

in which $Q^{ij} = g_{k\ell} \mathcal{P}^{ik} \mathcal{P}^{j\ell}$, and

$$\begin{aligned} \mathcal{P}_1 &= g_{ij} \mathcal{P}^{ij}, & \mathcal{P}_2 &= \frac{1}{2} (\mathcal{P}_1^2 - g_{ij} Q^{ij}), \\ \mathcal{P}_3 &= \frac{1}{3} (3\mathcal{P}_1 \mathcal{P}_2 - \mathcal{P}_1^3 + g_{ij} g_{k\ell} \mathcal{P}^{ik} Q^{j\ell}) \end{aligned} \tag{4.5}$$

are three principal invariants of \mathcal{P}^{ij} . Note that $\mathcal{P}^{ij} = 0$ only and only if $\varepsilon_{ij}^* = 0$.

According to (4.4), I_1, I_2, I_3 are expressed in the form of

$$\begin{aligned} I_1 &= \frac{1}{2} \mathcal{P}_1 + I_1 + J_1, \\ I_2 &= \frac{1}{4} \mathcal{P}_2 + \frac{1}{2} (\mathcal{P}_1 I_1 + \mathcal{P}_1 J_1 - J_1 - J_2) + J_4 - J_3 \\ &\quad + I_1 J_1 - I_1 J_2 - \mathcal{P}_2 I_2 - \mathcal{P}_1 J_3 + \mathcal{P}_1 I_1 J_1 + I_2, \\ I_3 &= \frac{1}{8} \mathcal{P}_3 + \frac{1}{4} (J_2 + \mathcal{P}_3 I_1 + \mathcal{P}_2 I_1 - \mathcal{P}_1 J_1) + \frac{1}{2} (J_3 - J_4 \\ &\quad + I_1 J_2 - I_1 J_1 + \mathcal{P}_1 I_2 + \mathcal{P}_3 I_2 + \mathcal{P}_1 J_3 - \mathcal{P}_1 I_1 J_1) \\ &\quad + \mathcal{P}_2 I_2 + (1 + \mathcal{P}_1 + \mathcal{P}_2 + \mathcal{P}_3) I_3, \end{aligned} \tag{4.6}$$

where J_1, J_2, J_3, J_4 are coupling terms concerning the influences of initial states to the subsequent deformations, i.e.,

$$J_1 = \mathcal{P}^{ij} \varepsilon_{ij}, \quad J_2 = Q^{ij} \varepsilon_{ij}, \quad J_3 = g^{ik} \mathcal{P}^{j\ell} \varepsilon_{ij} \varepsilon_{k\ell}, \quad J_4 = g^{ik} Q^{j\ell} \varepsilon_{ij} \varepsilon_{k\ell}. \tag{4.7}$$

Note that the following two identities

$$\begin{aligned} &(g^{j\ell} Q^{ik} + g^{i\ell} Q^{jk}) \varepsilon_{k\ell} + \mathcal{P}^{ik} \mathcal{P}^{j\ell} \varepsilon_{k\ell} \\ &= I_1 Q^{ij} - \mathcal{P}_2 I_1^{ij} + (J_2 + I_1 \mathcal{P}_2 - J_1 \mathcal{P}_1) g^{ij} + (J_1 - I_1 \mathcal{P}_1) \mathcal{P}^{ij} \\ &\quad + \mathcal{P}_1 (g^{j\ell} \mathcal{P}^{ik} + g^{i\ell} \mathcal{P}^{jk}) \varepsilon_{k\ell}, \end{aligned} \tag{4.8}$$

$$\begin{aligned} &g_{k\ell} (\mathcal{P}^{ik} I_2^{j\ell} + \mathcal{P}^{jk} I_2^{i\ell}) + g^{ik} g^{j\ell} \mathcal{P}^{mq} \varepsilon_{km} \varepsilon_{q\ell} \\ &= \mathcal{P}_1 I_2^{ij} - I_2 \mathcal{P}_1^{ij} + (J_3 - I_1 J_1 + I_2 \mathcal{P}_1) g^{ij} + (J_1 - I_1 \mathcal{P}_1) I_1^{ij} \\ &\quad + I_1 (g^{j\ell} \mathcal{P}^{ik} + g^{i\ell} \mathcal{P}^{jk}) \varepsilon_{k\ell} \end{aligned} \tag{4.9}$$

can be derived by repeatedly applying the Hamilton–Cayley theorem, thus the other four invariants can be expressed as the combination of (4.5) and (4.7)

$$\begin{aligned} \mathcal{P}^{ik} \mathcal{P}^{j\ell} \varepsilon_{ij} \varepsilon_{k\ell} &= 2I_1 J_2 + J_1^2 - 2J_4 + 2(J_3 - I_1 J_1) \mathcal{P}_1 + 2I_2 \mathcal{P}_2, \\ \mathcal{P}^{ik} Q^{j\ell} \varepsilon_{ij} \varepsilon_{k\ell} &= J_1 J_2 + [I_1 J_2 - J_4 + (J_3 - I_1 J_1) \mathcal{P}_1 + I_2 \mathcal{P}_2] \mathcal{P}_1 \\ &\quad + I_2 \mathcal{P}_3, \\ g^{ik} \mathcal{P}^{j\ell} \mathcal{P}^{mq} \varepsilon_{ij} \varepsilon_{km} \varepsilon_{q\ell} &= J_1 J_3 + I_1 (I_1 J_2 - J_4) + (I_1 J_3 - I_1^2 J_1) \mathcal{P}_1 \\ &\quad + (I_1 I_2 + I_3) \mathcal{P}_2, \\ \mathcal{P}^{ik} \mathcal{P}^{j\ell} \mathcal{P}^{mq} \varepsilon_{ij} \varepsilon_{km} \varepsilon_{q\ell} &= J_1 (J_1^2 + 3I_1 J_2 - 3J_4) + 3J_1 (J_3 - I_1 J_1) \mathcal{P}_1 \\ &\quad + 3(I_2 J_1 \mathcal{P}_2 + I_3 \mathcal{P}_3). \end{aligned} \tag{4.10}$$

These results demonstrate that the following ten independent invariants

$$I_1, I_2, I_3, \mathcal{P}_1, \mathcal{P}_2, \mathcal{P}_3, J_1, J_2, J_3, J_4 \tag{4.11}$$

compose a complete set of base function in describing the initially deformed elastomers (Fig. 2). The three invariants of (4.1) are equivalent to the ten invariants of (4.11). We use the latter since they show more clearly how the initial stress influences the subsequent deformation.

According to (2.3), the strain derivatives of I_1, I_2, I_3 can be calculated as

$$\frac{\partial I_1}{\partial \varepsilon_{ij}} = g^{ij}, \quad \frac{\partial I_2}{\partial \varepsilon_{ij}} = I_1 g^{ij} - I_1^{ij}, \quad \frac{\partial I_3}{\partial \varepsilon_{ij}} = I_2 g^{ij} - I_1 I_1^{ij} + I_2^{ij}. \tag{4.12}$$

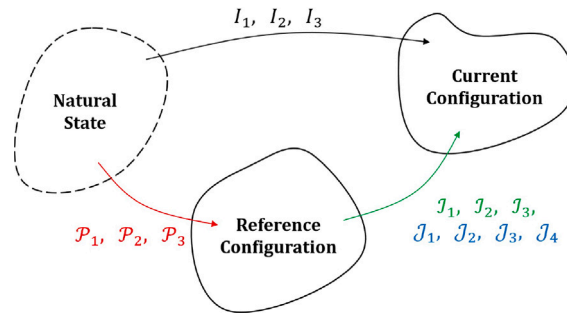


Fig. 2. Schematic diagram of the decomposition of invariants I_1, I_2, I_3 .

Similarly, according to (4.7), the strain derivatives of J_1, J_2, J_3, J_4 are

$$\frac{\partial J_1}{\partial \epsilon_{ij}} = P^{ij}, \quad \frac{\partial J_2}{\partial \epsilon_{ij}} = Q^{ij}, \quad \frac{\partial J_3}{\partial \epsilon_{ij}} = R^{ij}, \quad \frac{\partial J_4}{\partial \epsilon_{ij}} = S^{ij}, \tag{4.13}$$

where $R^{ij} = (g^{ik}P^{j\ell} + g^{jk}P^{i\ell}) \epsilon_{k\ell}$ and $S^{ij} = (g^{ik}Q^{j\ell} + g^{jk}Q^{i\ell}) \epsilon_{k\ell}$. Then by substituting (4.12) and (4.13) into (3.3), $\hat{\sigma}^{ij}$ can be derived as

$$\begin{aligned} \frac{\hat{\sigma}^{ij}}{\rho} = & \left\{ \left(\frac{\partial F}{\partial I_1} - \frac{p}{\rho} \frac{\partial C}{\partial I_1} \right) + (I_1 + J_1 - J_2 + J_1 P_1 - I_1 P_2 + \frac{1}{2} P_1) \times \right. \\ & \left(\frac{\partial F}{\partial I_2} - \frac{p}{\rho} \frac{\partial C}{\partial I_2} \right) + \frac{1}{4} \left[2(I_1 P_1 - J_1 P_1 + I_1 P_3 + J_2 - J_1) + P_2 \right. \\ & \left. \left. + P_3 + 4I_2(1 + P_1 + P_2 + P_3) + 4I_1 P_2 \right] \left(\frac{\partial F}{\partial I_3} - \frac{p}{\rho} \frac{\partial C}{\partial I_3} \right) \right\} g^{ij} \\ & + \left\{ (P_2 - 1) \left(\frac{\partial F}{\partial I_2} - \frac{p}{\rho} \frac{\partial C}{\partial I_2} \right) - \frac{1}{2} \left[2I_1(1 + P_1 + P_2 + P_3) \right. \right. \\ & \left. \left. + P_1 + 2P_2 + P_3 \right] \left(\frac{\partial F}{\partial I_3} - \frac{p}{\rho} \frac{\partial C}{\partial I_3} \right) \right\} I_1^{ij} \\ & + \left[\left(\frac{\partial F}{\partial I_1} - \frac{p}{\rho} \frac{\partial C}{\partial I_1} \right) + \frac{1}{2} (2I_1 + 2I_1 P_1 + P_1 - 1) \times \right. \\ & \left. \left(\frac{\partial F}{\partial I_2} - \frac{p}{\rho} \frac{\partial C}{\partial I_2} \right) - \frac{1}{4} (2I_1 P_1 + 2I_1 + P_1) \left(\frac{\partial F}{\partial I_3} - \frac{p}{\rho} \frac{\partial C}{\partial I_3} \right) \right] P^{ij} \\ & + \frac{1}{4} \left[\left(\frac{\partial F}{\partial I_3} - \frac{p}{\rho} \frac{\partial C}{\partial I_3} \right) - 2 \left(\frac{\partial F}{\partial I_2} - \frac{p}{\rho} \frac{\partial C}{\partial I_2} \right) \right] \left[(1 + 2I_1) Q^{ij} - 2S^{ij} \right. \\ & \left. + 2(1 + P_1) R^{ij} \right] + \left[(1 + P_1 + P_2 + P_3) \left(\frac{\partial F}{\partial I_3} - \frac{p}{\rho} \frac{\partial C}{\partial I_3} \right) \right] I_2^{ij}. \end{aligned} \tag{4.14}$$

If $C = P^{ij} = 0$, there is no internal constraint and initial deformation, thus $I_1 = I_1, I_2 = I_2, I_3 = I_3$, and (4.14) degenerate to the following classical forms

$$\frac{\hat{\sigma}^{ij}}{\rho} = \left(\frac{\partial F}{\partial I_1} + I_1 \frac{\partial F}{\partial I_2} + I_2 \frac{\partial F}{\partial I_3} \right) g^{ij} - \left(\frac{\partial F}{\partial I_2} + I_1 \frac{\partial F}{\partial I_3} \right) I_1^{ij} + \frac{\partial F}{\partial I_3} I_2^{ij}. \tag{4.15}$$

If $\epsilon_{ij} = 0$, there is no subsequent deformation, thus $I_1 = \frac{1}{2} P_1, I_2 = \frac{1}{4} P_2, I_3 = \frac{1}{8} P_3$, and (4.14) reduce to the following algebraic equations

$$\sigma_0^{ij} = \beta_0 g^{ij} + \beta_1 P^{ij} + \beta_2 Q^{ij}, \tag{4.16}$$

in which coefficients $\beta_0, \beta_1, \beta_2$ are explicitly expressed as

$$\begin{aligned} \beta_0 = & \frac{\rho_0}{4} \left[4 \left(\frac{\partial F}{\partial I_1} - \frac{p_0}{\rho_0} \frac{\partial C}{\partial I_1} \right) + 2P_1 \left(\frac{\partial F}{\partial I_2} - \frac{p_0}{\rho_0} \frac{\partial C}{\partial I_2} \right) \right. \\ & \left. + (P_2 + P_3) \left(\frac{\partial F}{\partial I_3} - \frac{p_0}{\rho_0} \frac{\partial C}{\partial I_3} \right) \right] \left(\frac{P_1}{2}, \frac{P_2}{4}, \frac{P_3}{8}, T_0, x^i \right), \\ \beta_1 = & \frac{\rho_0}{4} \left[4 \left(\frac{\partial F}{\partial I_1} - \frac{p_0}{\rho_0} \frac{\partial C}{\partial I_1} \right) + 2(P_1 - 1) \left(\frac{\partial F}{\partial I_2} - \frac{p_0}{\rho_0} \frac{\partial C}{\partial I_2} \right) \right. \\ & \left. - P_1 \left(\frac{\partial F}{\partial I_3} - \frac{p_0}{\rho_0} \frac{\partial C}{\partial I_3} \right) \right] \left(\frac{P_1}{2}, \frac{P_2}{4}, \frac{P_3}{8}, T_0, x^i \right), \\ \beta_2 = & \frac{\rho_0}{4} \left[\left(\frac{\partial F}{\partial I_3} - \frac{p_0}{\rho_0} \frac{\partial C}{\partial I_3} \right) - 2 \left(\frac{\partial F}{\partial I_2} - \frac{p_0}{\rho_0} \frac{\partial C}{\partial I_2} \right) \right] \left(\frac{P_1}{2}, \frac{P_2}{4}, \frac{P_3}{8}, T_0, x^i \right). \end{aligned} \tag{4.17}$$

By multiplying $g_{ik}\sigma_0^{k\ell}$ to both sides of (4.16), one obtains

$$g_{k\ell}\sigma_0^{ik}\sigma_0^{j\ell} = \alpha_0 g^{ij} + \alpha_1 \mathcal{P}^{ij} + \alpha_2 \mathcal{Q}^{ij}, \tag{4.18}$$

where coefficients $\alpha_0, \alpha_1, \alpha_2$ are derived as

$$\begin{aligned} \alpha_0 &= \beta_0^2 + 2\beta_1\beta_2\mathcal{P}_3 + \beta_2^2\mathcal{P}_1\mathcal{P}_3, \\ \alpha_1 &= 2\beta_0\beta_1 - 2\beta_1\beta_2\mathcal{P}_2 + \beta_2^2(\mathcal{P}_3 - \mathcal{P}_1\mathcal{P}_2), \\ \alpha_2 &= 2\beta_0\beta_2 + \beta_1^2 + 2\beta_1\beta_2\mathcal{P}_1 + \beta_2^2(\mathcal{P}_1^2 - \mathcal{P}_2). \end{aligned} \tag{4.19}$$

We then introduce the following three invariants of σ_0^{ij}

$$\mathcal{L}_1 = g_{ij}\sigma_0^{ij}, \quad \mathcal{L}_2 = g_{ij}g_{k\ell}\sigma_0^{ik}\sigma_0^{j\ell}, \quad \mathcal{L}_3 = g_{ij}g_{km}g_{q\ell}\sigma_0^{ik}\sigma_0^{j\ell}\sigma_0^{mq}. \tag{4.20}$$

By repeatedly employing the Hamilton–Cayley theorem to both (4.16) and (4.18), one obtains the following algebraic equations

$$\begin{aligned} \mathcal{L}_1 &= 3\beta_0 + \beta_1\mathcal{P}_1 + \beta_2(\mathcal{P}_1^2 - 2\mathcal{P}_2), \\ \mathcal{L}_2 &= 3\alpha_0 + \alpha_1\mathcal{P}_1 + \alpha_2(\mathcal{P}_1^2 - 2\mathcal{P}_2), \\ \mathcal{L}_3 &= 3\alpha_0\beta_0 + (\alpha_0\beta_1 + \alpha_1\beta_0)\mathcal{P}_1 \\ &\quad + (\alpha_0\beta_2 + \alpha_1\beta_1 + \alpha_2\beta_0)(\mathcal{P}_1^2 - 2\mathcal{P}_2) \\ &\quad + (\alpha_1\beta_2 + \alpha_2\beta_1)(\mathcal{P}_1^3 - 3\mathcal{P}_1\mathcal{P}_2 + 3\mathcal{P}_3) \\ &\quad + \alpha_2\beta_2(\mathcal{P}_1^4 - 4\mathcal{P}_1^2\mathcal{P}_2 + 4\mathcal{P}_1\mathcal{P}_3 + 2\mathcal{P}_2^2). \end{aligned} \tag{4.21}$$

To solve p_0 , we still need initial constraint equation

$$C\left(\frac{\mathcal{P}_1}{2}, \frac{\mathcal{P}_2}{4}, \frac{\mathcal{P}_3}{8}, T_0, x^i\right) = 0. \tag{4.22}$$

Simultaneous equations (4.21) and (4.22) are the isotropic versions of algebraic equations (3.5). Here, $\mathcal{P}_1, \mathcal{P}_2, \mathcal{P}_3, p_0$ are unknowns while $\mathcal{L}_1, \mathcal{L}_2, \mathcal{L}_3$ are given. The corresponding solutions take the form of

$$\begin{aligned} \mathcal{P}_1 &= \mathcal{P}_1(\rho_0, \mathcal{L}_1, \mathcal{L}_2, \mathcal{L}_3, \vartheta^*, T_0, x^i), \\ \mathcal{P}_2 &= \mathcal{P}_2(\rho_0, \mathcal{L}_1, \mathcal{L}_2, \mathcal{L}_3, \vartheta^*, T_0, x^i), \\ \mathcal{P}_3 &= \mathcal{P}_3(\rho_0, \mathcal{L}_1, \mathcal{L}_2, \mathcal{L}_3, \vartheta^*, T_0, x^i), \\ p_0 &= p_0(\rho_0, \mathcal{L}_1, \mathcal{L}_2, \mathcal{L}_3, \vartheta^*, T_0, x^i). \end{aligned} \tag{4.23}$$

One can solve (4.16) and (4.18) inversely and thus $\mathcal{P}^{ij}, \mathcal{Q}^{ij}$ become

$$\begin{aligned} \mathcal{P}^{ij} &= \frac{(\beta_2\alpha_0 - \beta_0\alpha_2)g^{ij} + \alpha_2\sigma_0^{ij} - \beta_2g_{k\ell}\sigma_0^{ik}\sigma_0^{j\ell}}{\beta_1\alpha_2 - \beta_2\alpha_1}, \\ \mathcal{Q}^{ij} &= \frac{(\beta_0\alpha_1 - \beta_1\alpha_0)g^{ij} - \alpha_1\sigma_0^{ij} + \beta_1g_{k\ell}\sigma_0^{ik}\sigma_0^{j\ell}}{\beta_1\alpha_2 - \beta_2\alpha_1}. \end{aligned} \tag{4.24}$$

We continue to introduce the coupling invariants of σ_0^{ij} and ε_{ij}

$$\begin{aligned} \mathcal{K}_1 &= \sigma_0^{ij}\varepsilon_{ij}, \quad \mathcal{K}_2 = g_{k\ell}\sigma_0^{ik}\sigma_0^{j\ell}\varepsilon_{ij}, \\ \mathcal{K}_3 &= g^{ik}\sigma_0^{j\ell}\varepsilon_{ij}\varepsilon_{k\ell}, \quad \mathcal{K}_4 = g^{ik}g_{mq}\sigma_0^{jm}\sigma_0^{q\ell}\varepsilon_{ij}\varepsilon_{k\ell}. \end{aligned} \tag{4.25}$$

Then by substituting (4.24) into (4.7), one obtains the following algebraic equations of four invariants

$$\begin{aligned} \mathcal{J}_1 &= \frac{(\beta_2\alpha_0 - \beta_0\alpha_2)\mathcal{I}_1 + \alpha_2\mathcal{K}_1 - \beta_2\mathcal{K}_2}{\beta_1\alpha_2 - \beta_2\alpha_1}, \\ \mathcal{J}_2 &= \frac{(\beta_0\alpha_1 - \beta_1\alpha_0)\mathcal{I}_1 - \alpha_1\mathcal{K}_1 + \beta_1\mathcal{K}_2}{\beta_1\alpha_2 - \beta_2\alpha_1}, \\ \mathcal{J}_3 &= \frac{(\beta_2\alpha_0 - \beta_0\alpha_2)(\mathcal{I}_1^2 - \mathcal{I}_2) + \alpha_2\mathcal{K}_3 - \beta_2\mathcal{K}_4}{\beta_1\alpha_2 - \beta_2\alpha_1}, \\ \mathcal{J}_4 &= \frac{(\beta_0\alpha_1 - \beta_1\alpha_0)(\mathcal{I}_1^2 - \mathcal{I}_2) - \alpha_1\mathcal{K}_3 + \beta_1\mathcal{K}_4}{\beta_1\alpha_2 - \beta_2\alpha_1}. \end{aligned} \tag{4.26}$$

Finally, by substituting (4.23) into (3.7), s_0 can be derived as

$$\begin{aligned} s_0 &= s_0(\rho_0, \mathcal{L}_1, \mathcal{L}_2, \mathcal{L}_3, \vartheta^*, T_0, x^i) \\ &= \left[\frac{p_0(\rho_0, \mathcal{L}_1, \mathcal{L}_2, \mathcal{L}_3, \vartheta^*, T_0, x^i)}{\rho_0} \frac{\partial C}{\partial \vartheta} - \frac{\partial \mathcal{F}}{\partial \vartheta} \right] \Big|_{\mathcal{P}_i = \mathcal{P}_i(\rho_0, \mathcal{L}_1, \mathcal{L}_2, \mathcal{L}_3, \vartheta^*, T_0, x^i)}. \end{aligned} \tag{4.27}$$

then from (4.23), (4.26) and (4.27), the equivalent free energy density and the equivalent internal constraint can be explicitly expressed as

$$\begin{aligned} \hat{F} &= \hat{F}(I_1, I_2, I_3, \mathcal{K}_1, \mathcal{K}_2, \mathcal{K}_3, \mathcal{K}_4, \vartheta, \rho_0, \mathcal{L}_1, \mathcal{L}_2, \mathcal{L}_3, \vartheta^*, T_0, x^i), \\ \hat{C} &= \hat{C}(I_1, I_2, I_3, \mathcal{K}_1, \mathcal{K}_2, \mathcal{K}_3, \mathcal{K}_4, \vartheta, \rho_0, \mathcal{L}_1, \mathcal{L}_2, \mathcal{L}_3, \vartheta^*, T_0, x^i), \end{aligned} \tag{4.28}$$

where $\rho_0, \mathcal{L}_1, \mathcal{L}_2, \mathcal{L}_3, \vartheta^*$, and T_0 serve as given parameters. According to (3.8) and (3.9), \hat{F} and \hat{C} are functional of $g_{ij}, \varepsilon_{ij}, \sigma_0^{ij}, \vartheta, \rho_0, \vartheta^*, T_0$, and x^i . These variables are undoubtedly independent. The difference between (3.8), (3.9), and (4.28) is that g_{ij}, ε_{ij} , and σ_0^{ij} are combined into ten invariants $I_1, I_2, I_3, \mathcal{L}_1, \mathcal{L}_2, \mathcal{L}_3, \mathcal{K}_1, \mathcal{K}_2, \mathcal{K}_3, \mathcal{K}_4$. According to the theory of representations for isotropic tensor functions (Zheng, 1994), the ten invariants are independent. Therefore, all the variables on the right side of (4.28) are independent.

5. Linearization

5.1. The general forms of the linearized constitutive equations

If displacement gradients and temperature variation are small enough (in comparison with the natural temperature) while other quantities remain finite, all the above derivations can be linearized to the first-order of $\nabla_i u_j$ and $\vartheta = T - T_0$. Geometric equations (2.8) now reduce to the classical Cauchy strains $\varepsilon_{ij} = \frac{1}{2} (\nabla_i u_j + \nabla_j u_i)$, then (2.1) become

$$G^{ij} = g^{ij} - 2I_1^{ij} = g^{ij} - g^{jk} \nabla_k u^i - g^{ik} \nabla_k u^j. \tag{5.1}$$

The square root of (2.4) is linearized as

$$\sqrt{G} = \sqrt{g} (1 + I_1) = \sqrt{g} (1 + \nabla_i u^i), \tag{5.2}$$

thus according to continuity Eq. (2.5), ρ degenerates to the classical form

$$\rho = \rho_0 (1 - I_1) = \rho_0 (1 - \nabla_i u^i). \tag{5.3}$$

Hoger and Johnson have demonstrated that linearizations cannot be performed before taking the respective derivatives (Hoger & Johnson, 1995a, 1995b). Thus equivalent internal constraint (3.9) should be expended to the second order of ε_{ij} and ϑ (Chadwick & Scott, 1992), i.e.,

$$\hat{C} = \chi^{ij} \varepsilon_{ij} - \xi \vartheta + \frac{1}{2} C^{ijkl} \varepsilon_{ij} \varepsilon_{kl} - \phi^{ij} \varepsilon_{ij} \vartheta - \frac{\eta}{2} \vartheta^2, \tag{5.4}$$

where the coefficients are derived in the form of

$$\begin{aligned} \chi^{ij} &= \left. \frac{\partial \hat{C}}{\partial \varepsilon_{ij}} \right|_0, & \xi &= - \left. \frac{\partial \hat{C}}{\partial \vartheta} \right|_0, & C^{ijkl} &= \left. \frac{\partial^2 \hat{C}}{\partial \varepsilon_{ij} \partial \varepsilon_{kl}} \right|_0, \\ \phi^{ij} &= - \left. \frac{\partial^2 \hat{C}}{\partial \varepsilon_{ij} \partial \vartheta} \right|_0, & \eta &= - \left. \frac{\partial^2 \hat{C}}{\partial \vartheta^2} \right|_0, \end{aligned} \tag{5.5}$$

in which “ $|_0$ ” means that it takes value at $\varepsilon_{ij} = \vartheta = 0$. The linearized constraint equation takes the following form

$$\chi^{ij} \varepsilon_{ij} = \chi^{ij} \nabla_i u_j = \xi \vartheta. \tag{5.6}$$

The physical explanation of ξ is that it represents the influences of temperature variation on the internal constraint (Humphrey & Rajagopal, 1997). If $\xi = 0$, (5.6) degenerates to the classical form of isothermal constraint $\chi^{ij} \nabla_i u_j = 0$. Similarly, equivalent free energy density (3.8) should also be expanded as

$$\hat{F} = \frac{p_0}{\rho_0} (\chi^{ij} \varepsilon_{ij} - \xi \vartheta) + \frac{1}{2} E^{ijkl} \varepsilon_{ij} \varepsilon_{kl} - \psi^{ij} \varepsilon_{ij} \vartheta - \frac{c}{2T_0} \vartheta^2, \tag{5.7}$$

where the coefficients are derived in the form of

$$E^{ijkl} = \left. \frac{\partial^2 \hat{F}}{\partial \varepsilon_{ij} \partial \varepsilon_{kl}} \right|_0, \quad \psi^{ij} = - \left. \frac{\partial^2 \hat{F}}{\partial \varepsilon_{ij} \partial \vartheta} \right|_0, \quad c = -T_0 \left. \frac{\partial^2 \hat{F}}{\partial \vartheta^2} \right|_0. \tag{5.8}$$

Here, c can be regarded as the heat capacity at constant reference volume.

By substituting (5.4) and (5.7) into (3.10), $\hat{\sigma}^{ij}$ are derived as

$$\begin{aligned} \hat{\sigma}^{ij} &= \sigma_0^{ij} + \left[\rho_0 E^{ijkl} - (p_0 + \hat{p}) C^{ijkl} - g^{kl} (\sigma_0^{ij} + p_0 \chi^{ij}) \right] \varepsilon_{kl} \\ &\quad + (p \phi^{ij} - \rho_0 \psi^{ij}) \vartheta - \chi^{ij} \hat{p}, \\ &= \sigma_0^{ij} + \left[\rho_0 E^{ijkl} - (p_0 + \hat{p}) C^{ijkl} \right] \nabla_k u_l - (\sigma_0^{ij} + p_0 \chi^{ij}) \nabla_k u^k \\ &\quad + (p \phi^{ij} - \rho_0 \psi^{ij}) \vartheta - \chi^{ij} \hat{p}, \end{aligned} \tag{5.9}$$

in which $\hat{p} = p - p_0$ is the increment of constraint multiplier, it may be large and thus bi-linear terms $\hat{p}\nabla_i u_j$ cannot be neglected. Then by substituting (5.9) into (2.11), Cauchy stresses σ^{ij} can be expressed as

$$\begin{aligned} \sigma^{ij} = & \sigma_0^{ij} + \left(\rho_0 E^{ijk\ell} + g^{i\ell} \sigma_0^{jk} + g^{j\ell} \sigma_0^{ik} \right) \nabla_k u_\ell \\ & - \left[(p_0 + \hat{p}) C^{ijk\ell} + \hat{p} (g^{i\ell} \chi^{jk} + g^{j\ell} \chi^{ik}) \right] \nabla_k u_\ell \\ & - \left(\sigma_0^{ij} + p_0 \chi^{ij} \right) \nabla_k u^k + \left[(p_0 + \hat{p}) \phi^{ij} - \rho_0 \psi^{ij} \right] \vartheta - \chi^{ij} \hat{p}. \end{aligned} \tag{5.10}$$

Also according to (3.10), entropy s can be expressed in the form of

$$\begin{aligned} s = & s_0 + \left[\psi^{ij} - \frac{p_0 + \hat{p}}{\rho_0} (\phi^{ij} + \xi g^{ij}) \right] \varepsilon_{ij} + \left[\frac{c}{T_0} - \frac{\eta (p_0 + \hat{p})}{\rho_0} \right] \vartheta - \frac{\xi \hat{p}}{\rho_0}, \\ = & s_0 + \left[\psi^{ij} - \frac{p_0 + \hat{p}}{\rho_0} (\phi^{ij} + \xi g^{ij}) \right] \nabla_i u_j + \left[\frac{c}{T_0} - \frac{\eta (p_0 + \hat{p})}{\rho_0} \right] \vartheta - \frac{\xi \hat{p}}{\rho_0}. \end{aligned} \tag{5.11}$$

(5.10) and (5.11) together constitute the linearized constitutive equations.

5.2. The naturally isotropic and temperature-controlled-compressible elastomer

In practice, the elastomer is often assumed to be naturally isotropic and temperature-controlled-compressible. To be specific, the internal constraint is specified in the following form (Gurtin & Guidugli, 1973)

$$C = \sqrt{G} - \sqrt{g^*} - \zeta(T) = 0, \tag{5.12}$$

where ζ is a given function satisfies $\zeta(T^*) = 0$, it means that the volume of the elastomer is controlled by temperatures, and if $T \equiv T^*$, i.e., isothermal deformations, thus (5.12) degenerates to classical incompressible constraint $G = g^*$. By substituting (2.4) into (5.12), one concludes that

$$\begin{aligned} \hat{C} = & \sqrt{g} \left(\sqrt{1 + 2I_1 + 4I_2 + 8I_3} - \frac{1}{\sqrt{1 + \mathcal{P}_1 + \mathcal{P}_2 + \mathcal{P}_3}} \right) - \zeta(T_0 + \vartheta), \\ = & \sqrt{g} \left(\sqrt{1 + 2I_1 + 4I_2 + 8I_3} - 1 \right) - \hat{\zeta}(T_0, \vartheta), \end{aligned} \tag{5.13}$$

where $\hat{\zeta}(T_0, \vartheta) = \zeta(T_0 + \vartheta) - \zeta(T_0)$ is the increment of ζ , and

$$\sqrt{g} - \sqrt{g^*} = \sqrt{g} \left(1 - \frac{1}{\sqrt{1 + \mathcal{P}_1 + \mathcal{P}_2 + \mathcal{P}_3}} \right) = \zeta(T_0) \tag{5.14}$$

is the explicit form of initial constraint Eq. (4.22). Then by substituting (5.13) into (5.4), \hat{C} can be expanded in the form of

$$\hat{C} = \sqrt{g} \left(I_1 + 2I_2 - \frac{1}{2} I_1^2 \right) - \frac{\partial \hat{\zeta}}{\partial \vartheta} \Big|_0 \vartheta - \frac{1}{2} \frac{\partial^2 \hat{\zeta}}{\partial \vartheta^2} \Big|_0 \vartheta^2. \tag{5.15}$$

According to (5.15), coefficients (5.5) are explicitly derived as

$$\begin{aligned} C^{ijk\ell} = & \sqrt{g} (g^{ij} g^{k\ell} - g^{ik} g^{j\ell} - g^{i\ell} g^{jk}), \\ \chi^{ij} = & \sqrt{g} g^{ij}, \quad \xi = \frac{\partial \hat{\zeta}}{\partial \vartheta} \Big|_0, \quad \phi^{ij} = 0, \quad \eta = \frac{\partial^2 \hat{\zeta}}{\partial \vartheta^2} \Big|_0. \end{aligned} \tag{5.16}$$

Thus linearized constraint Eq. (5.6) becomes

$$\nabla_i u^i = \frac{\xi \vartheta}{\sqrt{g}}. \tag{5.17}$$

If $\xi = 0$, (5.17) degenerates to classical incompressible condition $\nabla_i u^i = 0$.

Naturally isotropic free energy density (4.1) now can be expanded as

$$\begin{aligned} F = & \frac{1}{4} \left[4K_1 + 2K_2 \mathcal{P}_1 + K_3 (\mathcal{P}_2 + \mathcal{P}_3) \right] I_1 + \frac{1}{4} (K_3 - 2K_2) J_2 \\ & + \frac{1}{4} \left[4K_1 + 2K_2 (\mathcal{P}_1 - 1) - K_3 \mathcal{P}_1 \right] J_1 + K_4 \vartheta \\ & + \frac{1}{2} (K_3 - 2K_2) (1 + \mathcal{P}_1) J_3 + \frac{1}{2} (2K_2 - K_3) J_4 \\ & + \frac{1}{2} \left[2K_5 \mathcal{P}_1 + 2K_6 (\mathcal{P}_2 + \mathcal{P}_3) + 2K_7 \mathcal{P}_1 (\mathcal{P}_2 + \mathcal{P}_3) \right. \\ & \quad \left. + K_{11} + K_{12} \mathcal{P}_1^2 + K_{13} (\mathcal{P}_2 + \mathcal{P}_3)^2 \right] I_1^2 \\ & + \frac{1}{2} \left[2K_2 (1 - \mathcal{P}_2) + K_3 (\mathcal{P}_1 + 2\mathcal{P}_2 + \mathcal{P}_3) \right] I_2 \\ & + \frac{1}{2} \left[2K_5 (\mathcal{P}_1 - 1) - 2K_6 \mathcal{P}_1 - 2K_7 \mathcal{P}_1 (\mathcal{P}_1 - 1) \right] \end{aligned}$$

$$\begin{aligned}
 & + K_{11} + K_{12} (\mathcal{P}_1 - 1)^2 + K_{13} \mathcal{P}_1^2 \Big] \mathcal{J}_1^2 \\
 & + \frac{1}{2} \Big[(2K_2 - K_3) (1 + \mathcal{P}_1) + 2K_5 (2\mathcal{P}_1 - 1) - 2K_7 \mathcal{P}_1^2 \\
 & \quad + 2K_6 (\mathcal{P}_2 + \mathcal{P}_3 - \mathcal{P}_1) + 2K_7 (\mathcal{P}_1 - 1) (\mathcal{P}_2 + \mathcal{P}_3) \\
 & \quad + 2K_{11} + 2K_{12} \mathcal{P}_1 (\mathcal{P}_1 - 1) - 2K_{13} \mathcal{P}_1 (\mathcal{P}_2 + \mathcal{P}_3) \Big] \mathcal{I}_1 \mathcal{J}_1 \\
 & + \frac{1}{2} \Big[K_3 - 2K_2 - 2K_5 + 2K_6 + 2K_7 (\mathcal{P}_1 - \mathcal{P}_2 - \mathcal{P}_3) \\
 & \quad - 2K_{12} \mathcal{P}_1 + 2K_{13} (\mathcal{P}_2 + \mathcal{P}_3) \Big] \mathcal{I}_1 \mathcal{J}_2 \\
 & + \Big[K_6 - K_5 + K_7 (2\mathcal{P}_1 - 1) - K_{12} (\mathcal{P}_1 - 1) - K_{13} \mathcal{P}_1 \Big] \mathcal{J}_1 \mathcal{J}_2 \\
 & + \frac{1}{2} (K_{12} - 2K_7 + K_{13}) \mathcal{J}_2^2 + \Big[K_8 + K_9 \mathcal{P}_1 + K_{10} (\mathcal{P}_2 + \mathcal{P}_3) \Big] \mathcal{I}_1 \vartheta \\
 & + \Big[K_8 + K_9 (\mathcal{P}_1 - 1) - K_{10} \mathcal{P}_1 \Big] \mathcal{J}_1 \vartheta + (K_{10} - K_9) \mathcal{J}_2 \vartheta + \frac{1}{2} K_{14} \vartheta^2,
 \end{aligned} \tag{5.18}$$

in which the coefficients are expressed as

$$\begin{aligned}
 K_1 &= \frac{\partial \mathcal{F}}{\partial I_1} \Big|_0, \quad K_2 = \frac{\partial \mathcal{F}}{\partial I_2} \Big|_0, \quad K_3 = \frac{\partial \mathcal{F}}{\partial I_3} \Big|_0, \quad K_4 = \frac{\partial \mathcal{F}}{\partial \vartheta} \Big|_0, \\
 K_5 &= \frac{1}{2} \frac{\partial^2 \mathcal{F}}{\partial I_1 \partial I_2} \Big|_0, \quad K_6 = \frac{1}{4} \frac{\partial^2 \mathcal{F}}{\partial I_1 \partial I_3} \Big|_0, \quad K_7 = \frac{1}{8} \frac{\partial^2 \mathcal{F}}{\partial I_2 \partial I_3} \Big|_0, \\
 K_8 &= \frac{\partial^2 \mathcal{F}}{\partial I_1 \partial \vartheta} \Big|_0, \quad K_9 = \frac{1}{2} \frac{\partial^2 \mathcal{F}}{\partial I_2 \partial \vartheta} \Big|_0, \quad K_{10} = \frac{1}{4} \frac{\partial^2 \mathcal{F}}{\partial I_3 \partial \vartheta} \Big|_0, \\
 K_{11} &= \frac{\partial^2 \mathcal{F}}{\partial I_1^2} \Big|_0, \quad K_{12} = \frac{1}{4} \frac{\partial^2 \mathcal{F}}{\partial I_2^2} \Big|_0, \quad K_{13} = \frac{1}{16} \frac{\partial^2 \mathcal{F}}{\partial I_3^2} \Big|_0, \quad K_{14} = \frac{\partial^2 \mathcal{F}}{\partial \vartheta^2} \Big|_0.
 \end{aligned} \tag{5.19}$$

Therefore, elastic coefficients E^{ijkl} are derived in the following forms

$$\begin{aligned}
 E^{ijkl} &= \left\{ K_2 (1 - \mathcal{P}_2) + \frac{K_3}{2} (\mathcal{P}_1 + 2\mathcal{P}_2 + \mathcal{P}_3) + 2K_5 \mathcal{P}_1 + K_{12} \mathcal{P}_1^2 \right. \\
 & \quad \left. + K_{11} + (\mathcal{P}_2 + \mathcal{P}_3) [2K_6 + 2K_7 \mathcal{P}_1 + K_{13} (\mathcal{P}_2 + \mathcal{P}_3)] \right\} g^{ij} g^{kl} \\
 & + \left[\frac{K_2}{2} (\mathcal{P}_2 - 1) - \frac{K_3}{4} (\mathcal{P}_1 + 2\mathcal{P}_2 + \mathcal{P}_3) \right] (g^{ik} g^{jl} + g^{jk} g^{il}) \\
 & + \left[\frac{1}{2} (2K_2 - K_3) (1 + \mathcal{P}_1) + K_5 (2\mathcal{P}_1 - 1) - K_7 \mathcal{P}_1^2 \right. \\
 & \quad \left. + K_{11} + K_6 (\mathcal{P}_2 + \mathcal{P}_3 - \mathcal{P}_1) + K_7 (\mathcal{P}_2 + \mathcal{P}_3) (\mathcal{P}_1 - 1) \right. \\
 & \quad \left. + K_{12} \mathcal{P}_1 (\mathcal{P}_1 - 1) - K_{13} (\mathcal{P}_2 + \mathcal{P}_3) \mathcal{P}_1 \right] (g^{ij} \mathcal{P}^{kl} + g^{kl} \mathcal{P}^{ij}) \\
 & + \left[\frac{K_3}{2} - K_2 - K_5 + K_6 + K_7 (\mathcal{P}_1 - \mathcal{P}_2 - \mathcal{P}_3) \right. \\
 & \quad \left. - K_{12} \mathcal{P}_1 + K_{13} (\mathcal{P}_2 + \mathcal{P}_3) \right] (g^{ij} \mathcal{Q}^{kl} + g^{kl} \mathcal{Q}^{ij}) \\
 & + \left[2K_5 (\mathcal{P}_1 - 1) - 2K_6 \mathcal{P}_1 + 2K_7 \mathcal{P}_1 (1 - \mathcal{P}_1) \right. \\
 & \quad \left. + K_{11} + K_{12} (\mathcal{P}_1 - 1)^2 + K_{13} \mathcal{P}_1^2 \right] \mathcal{P}^{ij} \mathcal{P}^{kl} \\
 & + \left[K_6 - K_5 + K_7 (2\mathcal{P}_1 - 1) \right. \\
 & \quad \left. - K_{12} (\mathcal{P}_1 - 1) - K_{13} \mathcal{P}_1 \right] (\mathcal{P}^{ij} \mathcal{Q}^{kl} + \mathcal{P}^{kl} \mathcal{Q}^{ij}) \\
 & + \frac{1}{4} (K_3 - 2K_2) (\mathcal{P}_1 + 1) (g^{ik} \mathcal{P}^{jl} + g^{il} \mathcal{P}^{jk} + g^{jk} \mathcal{P}^{il} + g^{il} \mathcal{P}^{jk}) \\
 & + \frac{1}{4} (2K_2 - K_3) (g^{ik} \mathcal{Q}^{jl} + g^{il} \mathcal{Q}^{jk} + g^{jk} \mathcal{Q}^{il} + g^{il} \mathcal{Q}^{jk}) \\
 & + (K_{12} - 2K_7 + K_{13}) \mathcal{Q}^{ij} \mathcal{Q}^{kl}.
 \end{aligned} \tag{5.20}$$

Also coupling coefficients ψ^{ij} are derived as

$$\begin{aligned}
 \psi^{ij} &= [K_{10} \mathcal{P}_1 + K_9 (1 - \mathcal{P}_1) - K_8] \mathcal{P}^{ij} + (K_9 - K_{10}) \mathcal{Q}^{ij} \\
 & - [K_8 + K_9 \mathcal{P}_1 + K_{10} (\mathcal{P}_2 + \mathcal{P}_3)] g^{ij}.
 \end{aligned} \tag{5.21}$$

Besides, $c = -T_0 K_{14}$. Algebraic equations (4.16) now become

$$\begin{aligned} \sigma_0^{ij} &= \frac{\rho_0}{4} \left[4K_1 + 2K_2 \mathcal{P}_1 + K_3 (\mathcal{P}_2 + \mathcal{P}_3) - 4\sqrt{g} \frac{\rho_0}{\rho_0} \right] g^{ij} \\ &+ \frac{\rho_0}{4} [4K_1 + 2K_2 (\mathcal{P}_1 - 1) - K_3 \mathcal{P}_1] \mathcal{P}^{ij} + \frac{\rho_0}{4} (K_3 - 2K_2) Q^{ij}, \end{aligned} \tag{5.22}$$

while initial entropy (4.27) becomes

$$s_0 = -K_4 - \frac{\rho_0}{\rho_0} \xi. \tag{5.23}$$

Finally, by substituting (5.16) and (5.20) into (5.10), σ^{ij} are derived as

$$\begin{aligned} \sigma^{ij} &= \sigma_0^{ij} + \left[\rho_0 E^{ijk\ell} + \sqrt{g} \rho_0 (g^{ik} g^{j\ell} + g^{i\ell} g^{jk}) + g^{i\ell} \sigma_0^{jk} + g^{j\ell} \sigma_0^{ik} \right] \nabla_k u_\ell \\ &- \left[\left(\frac{\sigma_0^{ij}}{\sqrt{g}} + 2\rho_0 g^{ij} + \hat{p} g^{ij} \right) \xi + \rho_0 \psi^{ij} \right] \vartheta - \sqrt{g} g^{ij} \hat{p}. \end{aligned} \tag{5.24}$$

Also by substituting (5.16) and (5.20) into (5.11), s can be derived as

$$s = s_0 + \psi^{ij} \nabla_i u_j + \left[\frac{c}{T_0} - \frac{\eta (\rho_0 + \hat{p})}{\rho_0} - \frac{(\rho_0 + \hat{p}) \xi^2}{\rho_0 \sqrt{g}} \right] \vartheta - \frac{\xi \hat{p}}{\rho_0}. \tag{5.25}$$

According to (5.24) and (5.25), one concludes that if $\xi, \eta \neq 0$, bi-linear term $\hat{p}\vartheta$ cannot be eliminated and the corresponding linearized equations are still somewhat nonlinear. Further assume that σ_0^{ij} are small compared with the elastic moduli of the elastomer while $\xi, \eta = 0$, then all the terms containing $\sigma_0^{ij} \nabla_k u_\ell, \rho_0 \nabla_i u_j$ or $\sigma_0^{ij} \vartheta$ are higher-order infinitesimal, thus can be neglected. By this means, (5.24) and (5.25) degenerate to

$$\begin{aligned} \sigma^{ij} &= \sigma_0^{ij} - \frac{\rho_0 K_2}{2} (g^{ik} g^{j\ell} + g^{jk} g^{i\ell}) \nabla_k u_\ell + \rho_0 K_8 g^{ij} \vartheta - \sqrt{g} g^{ij} \hat{p}, \\ s &= s_0 - K_{14} \vartheta, \end{aligned} \tag{5.26}$$

which are the classical constitutive equations of constrained thermoelastic elastomers with initial stresses (Kachanov, Shafiro, & Tsukrov, 2003). Here, only three coefficients K_2, K_8, K_{14} are required, and the superposition principle is valid.

6. Examples

6.1. The Saint Venant–Kirchhoff elastomer

In the first example, we aim to provide thermo-mechanically coupled constitutive equations for a one-dimensional Saint Venant–Kirchhoff elastomer with arbitrary initial states (it deforms in one direction, and the deformations in the other two directions are assumed to be zero, similar to plane waves). Here, the aforementioned embedding method can be carried out explicitly and clearly while the concrete usage of our new formulations is illustrated in detail.

The only material coordinate is x , thus all the tensor indices can be omitted, i.e., $g_{xx}^*, g_{xx}, G_{xx}, \sigma_0^{xx}, \sigma^{xx}, \epsilon_{xx}^*, \epsilon_{xx}, \mathcal{P}^{xx}$ are denoted by $g^*, g, G, \sigma_0, \sigma, \epsilon$ and \mathcal{P} , respectively. If $B_0 = B^*$, the free energy density of Saint Venant–Kirchhoff elastomer can be specified in the following quadratic form

$$\rho_0 F = \frac{\lambda}{2} I_1^2 + \mu I_2 - K \alpha I_1 (T - T_0) - \frac{\rho_0 c_v}{2T_0} (T - T_0)^2, \tag{6.1}$$

where λ, μ, K are Lamé constants and bulk modulus, respectively, α is the coefficient of bulk expansion and c_v is the heat capacity at constant reference volume. However, if $B_0 \neq B^*$, then B_0 is the reference configuration with arbitrary initial states while B^* is undetermined. (6.1) should be modified as

$$\rho^* F = \frac{\lambda}{2} I_1^2 + \mu I_2 - K \alpha I_1 (T - T^*) - \frac{\rho^* c_v}{2T^*} (T - T^*)^2. \tag{6.2}$$

For simplicity, g is assumed to be 1, thus $g^* = (1 + \mathcal{P})^{-1}$ according to (4.3). By substituting $g^* = 1 - 2\epsilon^*$ into (4.4), one concludes that

$$\mathcal{P} = \frac{2\epsilon^*}{1 - 2\epsilon^*}, \quad \epsilon^* = \frac{\mathcal{P}}{2(1 + \mathcal{P})}. \tag{6.3}$$

In this way, I_1 and I_2 can be derived as

$$I_1 = \frac{\epsilon^* + \epsilon}{g^*} = \frac{\epsilon^* + \epsilon}{1 - 2\epsilon^*} = \frac{\mathcal{P}}{2} + (1 + \mathcal{P}) \epsilon, \quad I_2 = 0. \tag{6.4}$$

Then according to $\rho^* \sqrt{g^*} = \rho_0$, (6.2) becomes

$$F = \frac{\mathcal{P} + 2(1 + \mathcal{P}) \epsilon}{8\rho_0 \sqrt{1 + \mathcal{P}}} \left[\lambda \mathcal{P} + 2\lambda (1 + \mathcal{P}) \epsilon - 4K \alpha (\vartheta^* + \vartheta) \right] - \frac{c_v (\vartheta^* + \vartheta)^2}{2T^*}. \tag{6.5}$$

By substituting (6.5) into (3.3), $\hat{\sigma}$ and s can be derived as

$$\begin{aligned} \frac{\hat{\sigma}}{\rho} &= \frac{\sqrt{1+\mathcal{P}}}{2\rho_0} [\lambda\mathcal{P} + 2\lambda(1+\mathcal{P})\varepsilon - 2K\alpha(\vartheta^* + \vartheta)], \\ s &= \frac{K\alpha[\mathcal{P} + 2(1+\mathcal{P})\varepsilon]}{2\rho_0\sqrt{1+\mathcal{P}}} + \frac{c_v(\vartheta^* + \vartheta)}{T^*}. \end{aligned} \tag{6.6}$$

From (3.4), one obtains $s^* = 0$. Then by substituting $\varepsilon = \vartheta = 0$ and $\rho = \rho_0$ into (6.6), algebraic equation (3.5)₁ now becomes

$$\sigma_0 = \frac{\sqrt{1+\mathcal{P}}}{2} (\lambda\mathcal{P} - 2K\alpha\vartheta^*). \tag{6.7}$$

Also initial entropy s_0 can be derived as

$$s_0 = \frac{K\alpha\mathcal{P}}{2\rho_0\sqrt{1+\mathcal{P}}} + \frac{c_v\vartheta^*}{T^*}. \tag{6.8}$$

Once σ_0, ϑ^* and dimensionless coefficients $K\alpha T^*/\lambda, \rho_0 c_v/K\alpha$ are given, (6.7) becomes a cubic equation of \mathcal{P} . If the solution of (6.7) exists, then by substituting this solution into (6.8), initial entropy s_0 can be accordingly obtained.

In many applications, (6.7) can be simplified. Firstly, the initial states are assumed to be originated from initially isothermal tensions/compressions, i.e., $\vartheta^* = 0$, then (6.7) and (6.8) become

$$\frac{\sigma_0}{\lambda} = \frac{\mathcal{P}}{2} \sqrt{1+\mathcal{P}}, \quad \frac{\rho_0 s_0}{K\alpha} = \frac{\mathcal{P}}{2\sqrt{1+\mathcal{P}}}. \tag{6.9}$$

To guarantee the existence of solutions of (6.9), a restriction on the possible values of σ_0 arises, that is $\sigma_0 \geq -(\sqrt{3}/9)\lambda$. This inequality indicates that there exists a minimum value of initial compression, which is similar to the buckling of rods. If the given σ_0 is smaller than $-(\sqrt{3}/9)\lambda$, (6.1) may no longer be an appropriate description of the elastomer. Furthermore, if $-(\sqrt{3}/9)\lambda \leq \sigma_0 < 0$, there exist two negative solutions, while if $\sigma_0 \geq 0$, only one positive solution exists. We here choose the branch that satisfies $\mathcal{P}(0, 0) = 0$.

Secondly, the initial states are assumed to be originated from initially free thermal expansions. By substituting $\sigma_0 = 0$ into (6.7), one concludes that

$$\mathcal{P} = \frac{2K\alpha T^*}{\lambda} \frac{\vartheta^*}{T^*}, \quad \frac{\rho_0 s_0}{K\alpha} = \frac{\mathcal{P}}{2\sqrt{1+\mathcal{P}}} + \frac{\rho_0 c_v}{K\alpha} \frac{\vartheta^*}{T^*}. \tag{6.10}$$

Once ϑ^* is given, then both \mathcal{P} and s_0 can be uniquely determined. Lastly, the initial states are assumed to be originated from initially adiabatic tensions/compressions. By substituting $s_0 = s^* = 0$ into (6.8), one obtains

$$\frac{\sigma_0}{\lambda} = \frac{\sqrt{1+\mathcal{P}}}{2} \left(\mathcal{P} - 2 \frac{K\alpha T^*}{\lambda} \frac{\vartheta^*}{T^*} \right), \quad \frac{\mathcal{P}}{2\sqrt{1+\mathcal{P}}} = - \frac{\rho_0 c_v}{K\alpha} \frac{\vartheta^*}{T^*}. \tag{6.11}$$

In (6.11), σ_0 and ϑ^* cannot be given independently. If one gives ϑ^* , then \mathcal{P} and σ_0 can be determined uniquely. While if one gives σ_0 , (6.11) reduce to another cubic equation that is different from (6.9). This fact indicates that the classical approaches, which concern only mechanical but not thermal effects, may be insufficient to derive the appropriate constitutive equations. Different thermo-mechanically coupled deformation histories correspond to different subsequent mechanical responses, even if they share the same σ_0 . In particular, when \mathcal{P} and ϑ^* are small, (6.7) and (6.8) reduce to linear equations

$$\frac{\sigma_0}{\lambda} = \frac{\mathcal{P}}{2} - \frac{K\alpha T^*}{\lambda} \frac{\vartheta^*}{T^*}, \quad \frac{\rho_0 s_0}{K\alpha} = \frac{\mathcal{P}}{2} + \frac{\rho_0 c_v}{K\alpha} \frac{\vartheta^*}{T^*}. \tag{6.12}$$

The solutions of (6.12) are expressed as

$$\begin{aligned} \mathcal{P}_{\text{lin}} &= 2 \left(\frac{\sigma_0}{\lambda} + \frac{K\alpha T^*}{\lambda} \frac{\vartheta^*}{T^*} \right), \\ \left(\frac{\rho_0 s_0}{K\alpha} \right)_{\text{lin}} &= \frac{\sigma_0}{\lambda} + \left(\frac{K\alpha T^*}{\lambda} + \frac{\rho_0 c_v}{K\alpha} \right) \frac{\vartheta^*}{T^*}. \end{aligned} \tag{6.13}$$

Now we introduce two dimensionless coefficients $\gamma = \sqrt{1+\mathcal{P}}$ and $\beta = T_0/T^*$ to replace \mathcal{P} and ϑ^* , respectively. The equivalent free energy density becomes

$$\hat{F} = \frac{\gamma^3 \lambda}{2\rho_0} \varepsilon^2 - \frac{\gamma K\alpha}{\rho_0} \varepsilon \vartheta - \frac{\beta c_v}{2T_0} \vartheta^2. \tag{6.14}$$

According to (6.14), one concludes that

$$E^{\text{xxxx}} = \frac{\gamma^3 \lambda}{\rho_0}, \quad \psi^{\text{xx}} = \frac{\gamma K\alpha}{\rho_0}, \quad c = \beta c_v. \tag{6.15}$$

By substituting (6.15) into (5.10) and (5.11), the thermo-mechanically coupled constitutive equations of one-dimensional Saint Venant–Kirchhoff elastomers with arbitrary initial states are derived in the form of

$$\begin{aligned} \sigma &= \sigma_0 + (\lambda\gamma^3 + \sigma_0)\varepsilon - \gamma K\alpha\vartheta = \sigma_0 + \lambda'\varepsilon - K'\alpha'\vartheta, \\ s &= s_0 + \frac{\gamma K\alpha}{\rho_0}\varepsilon + \frac{\beta c_v}{T_0}\vartheta = s_0 + \frac{K'\alpha'}{\rho_0}\varepsilon + \frac{c'_v}{T_0}\vartheta, \end{aligned} \tag{6.16}$$

where

$$\frac{\lambda'}{\lambda} = \frac{K'}{K} = \gamma^3 + \frac{\sigma_0}{\lambda}, \quad \frac{\alpha'}{\alpha} = \frac{\gamma}{\gamma^3 + \frac{\sigma_0}{\lambda}}, \quad \frac{c'_v}{c_v} = \beta \tag{6.17}$$

are ratios between the initial-states-effected values and their natural ones. The influences of initial states are reflected in coefficients γ and β . If $\sigma_0 = \vartheta^* = 0$, then $\gamma = \beta = 1$, thus $\lambda' = \lambda, K' = K, \alpha' = \alpha, c'_v = c_v$ and (6.16) reduce to

$$\sigma = \sigma_0 + \lambda\varepsilon - K\alpha\vartheta, \quad s = s_0 + \frac{K\alpha}{\rho_0}\varepsilon + \frac{c_v}{T_0}\vartheta. \tag{6.18}$$

To illustrate the quantitative influences of initial states, we here take nylon, for instance. Here, T^* is assumed to be 300 K, and coefficients $K\alpha T^*/\lambda$ and $\rho_0 c_v/K\alpha$ are specified as 0.076 and 2.2, respectively. We first consider initially isothermal tension/compression (6.9), where $\sigma_0/\lambda = \gamma(\gamma^2 - 1)/2$ and $\rho_0 s_0/K\alpha = (\gamma^2 - 1)/2\gamma$. The relations between various coefficients and σ_0/λ are plotted in Fig. 3(a). It can be concluded that $\lambda' > \lambda, K' > K, \alpha' < \alpha$ and $s_0 > 0$ when $\sigma_0 > 0$, but quite the opposite when $\sigma_0 < 0$. The heat capacity remains unchanged because $\beta \equiv 1$. We then consider initially free thermal expansion (6.10), where $\lambda'/\lambda = K'/K = \gamma^3$ and $\alpha'/\alpha = \gamma^{-2}$. The relations between various coefficients and $\beta = 1 + (\vartheta^*/T^*)$ are plotted in Fig. 3(b). It can be concluded that $\lambda' > \lambda, K' > K, c'_v > c_v, \alpha' < \alpha$ and $s_0 > 0$ when $T_0 > T^*$, but quite the opposite when $T_0 < T^*$. We finally consider initially adiabatic tension/compression (6.11), where σ_0 is given. The relations between various coefficients and σ_0/λ are plotted in Fig. 3(c). In this situation, the mechanical coefficients, i.e., $\lambda'/\lambda, K'/K, \alpha'/\alpha$ and γ , behave in a similar way to those in the isothermal case, only small differences can be found between the isothermal and the adiabatic coefficients. However, the thermal coefficients, i.e., c'_v/c_v and β , behave in a completely different way to those in the isothermal case. Here, $T_0 < T^*$ and $c'_v < c_v$ when $\sigma_0 > 0$, but quite the opposite when $\sigma_0 < 0$.

These results prove that the constitutive coefficients may behave differently even for the same initial stresses if the latter is originated from isothermal and adiabatic deformations, respectively. Especially for the thermal coefficients, which are most influenced by thermo-mechanically coupled deformation histories. As a consequence, the subsequent thermoelastic behaviors could be completely distinct. From this perspective, our new proposed framework is necessary and has certain practical value.

Moreover, we would like to compare our additive-decomposition-based approach to the multiplicative-decomposition-based isothermal theory. Suppose the stress-free length of the elastomer is L^* , while the initial and the current length are L_0 and L , respectively. According to deformation gradients

$$\frac{L_0}{L^*} = \gamma_0, \quad \frac{L}{L_0} = \gamma_e, \quad \frac{L}{L^*} = \gamma_t, \tag{6.19}$$

one obtains classical multiplicative decomposition $\gamma_t = \gamma_0\gamma_e$. If the Cauchy stress is specified in the form of $\sigma = \Sigma(\gamma_t)$, which satisfies $\Sigma(1) = 0$, then

$$\sigma_0 = \Sigma(\gamma_0), \quad \sigma = \Sigma(\gamma_0\gamma_e). \tag{6.20}$$

Now, we introduce the following Green strain

$$\varepsilon_e = \frac{1}{2} \left(\frac{L^2 - L_0^2}{L_0^2} \right) = \frac{1}{2} (\gamma_e^2 - 1), \quad \gamma_e = \sqrt{1 + 2\varepsilon_e}. \tag{6.21}$$

When ε_e is small, the Cauchy stress is derived in the form of

$$\sigma = \Sigma(\gamma_0 + \gamma_0\varepsilon_e) = \sigma_0 + \gamma_0 \frac{\partial \Sigma(\gamma_0)}{\partial \gamma_t} \varepsilon_e. \tag{6.22}$$

Then according to (6.6), one concludes that

$$\Sigma(\hat{\gamma}) = \frac{\lambda}{2} \gamma_t (\gamma_t^2 - 1), \quad \sigma_0 = \frac{\lambda}{2} \gamma_0 (\gamma_0^2 - 1), \quad \frac{\partial \Sigma(\gamma_0)}{\partial \gamma_t} = \frac{\lambda}{2} (3\gamma_0^2 - 1). \tag{6.23}$$

By substituting (6.23) into (6.22), one obtains the Cauchy stress

$$\sigma = \frac{\lambda}{2} \gamma_0 [\gamma_0^2 - 1 + (3\gamma_0^2 - 1)\varepsilon_e], \tag{6.24}$$

which is the same as (6.16), only and only if $\gamma_0 = \gamma, \varepsilon_e = \varepsilon$ and $\vartheta = 0$. This result verifies the equivalence of two approaches in the isothermal situations.

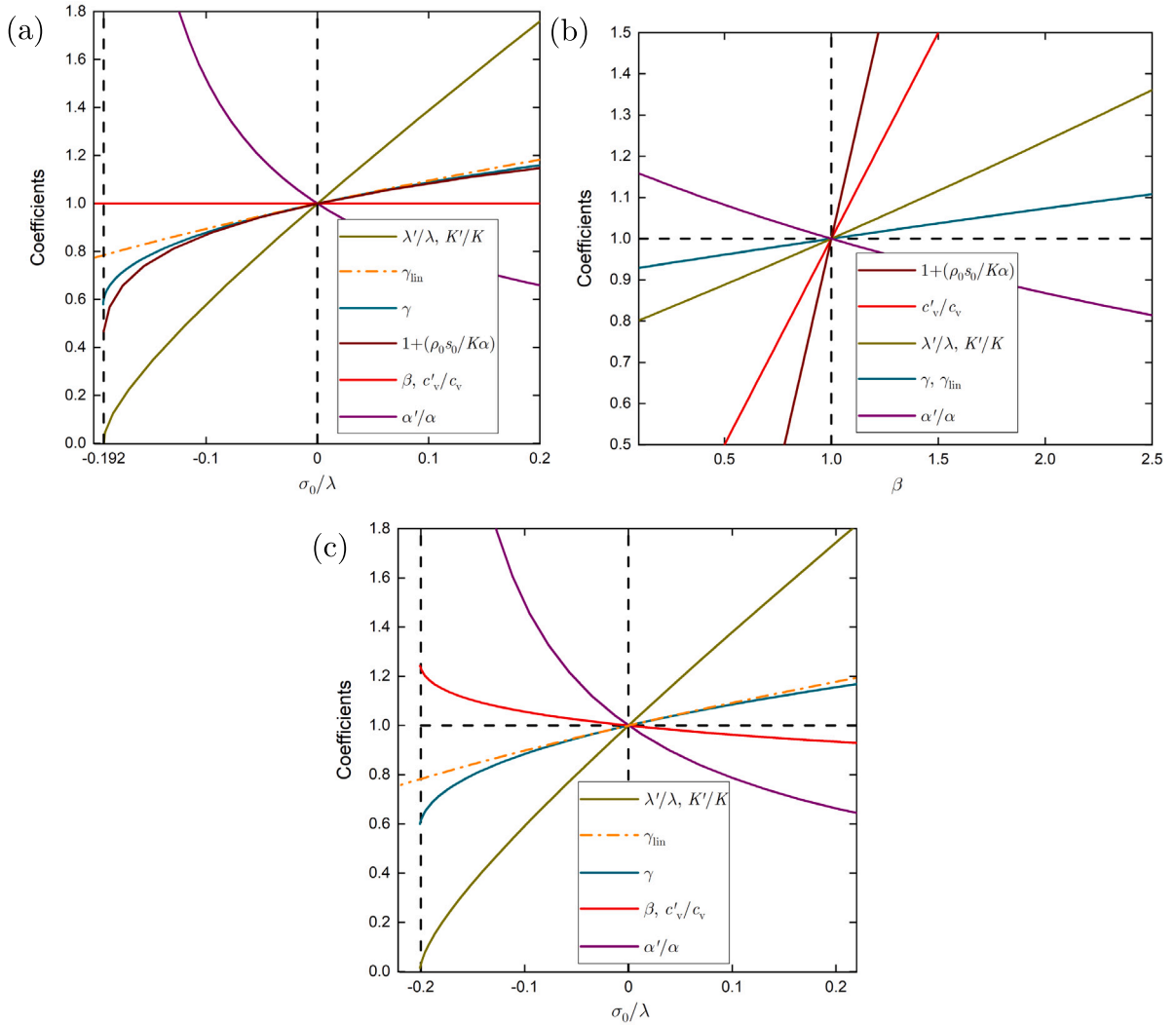


Fig. 3. Various coefficients vs dimensionless initial states that originated from different thermo-mechanically coupled deformation histories. Dotted line represents $\gamma_{lin} = \sqrt{1 + \mathcal{P}_{lin}^-}$. (a) Initially isothermal tension/compression (b) Initially free thermal expansion (c) Initially adiabatic tension/compression.

6.2. The Blatz–Ko elastomer

In the second example, we will study the three-dimensional deformations of Blatz–Ko elastomer, which has no internal constraint either. Suppose the initial states of the elastomer are originated from initially isothermal uniform tensions/compressions. Instead of I_1, I_2, I_3 , strain energies are often expressed in terms of the following three invariants in hyperelastic theory

$$\begin{aligned}
 \mathcal{G}_1 &= g^{*ij} G_{ij} = 3 + 2I_1, \\
 \mathcal{G}_2 &= \frac{1}{2} (\mathcal{G}_1^2 - g^{*ik} g^{*j\ell} G_{ij} G_{k\ell}) = 3 + 4I_1 + 4I_2, \\
 \mathcal{G}_3 &= \frac{1}{3} (3\mathcal{G}_1\mathcal{G}_2 - \mathcal{G}_1^3 + g^{*ik} g^{*j\ell} g^{*mq} G_{ij} G_{km} G_{q\ell}) = 1 + 2I_1 + 4I_2 + 8I_3.
 \end{aligned}
 \tag{6.25}$$

The natural strain energy density of Blatz–Ko elastomers is often specified as

$$\rho^* \mathcal{F} = \frac{\mu}{2} \left(\frac{\mathcal{G}_2}{\mathcal{G}_3} + 2\sqrt{\mathcal{G}_3} - 5 \right),
 \tag{6.26}$$

where μ is the elastic modulus. According to initial continuity equation $\rho_0\sqrt{g} = \rho^*\sqrt{g^*}$ and (6.25), the free energy density of Blatz–Ko elastomer becomes

$$F = \frac{\mu \left(\frac{3 + 4I_1 + 4I_2}{1 + 2I_1 + 4I_2 + 8I_3} + 2\sqrt{1 + 2I_1 + 4I_2 + 8I_3} \right)}{2\rho_0\sqrt{1 + P_1 + P_2 + P_3}}. \tag{6.27}$$

Here, x^i are specified as Descartes coordinates, where $g_{ij} = \delta_{ij}$ and thus there is no need to distinguish superscripts and subscripts. To initially uniform extension, $P^{ij} = P\delta^{ij}$ and thus $P_1 = 3P, P_2 = 3P^2, P_3 = P^3$. By introducing coefficient $\gamma = \sqrt{1 + P}$, then nine nonzero coefficients in (5.19) are derived as

$$\begin{aligned} K_1 &= \frac{\mu(2\gamma^2 + \gamma^5 - 3)}{\rho_0\gamma^{11}}, & K_2 &= \frac{2\mu(\gamma^2 + \gamma^5 - 3)}{\rho_0\gamma^{11}}, & K_3 &= \frac{4\mu(\gamma^5 - 3)}{\rho_0\gamma^{11}}, \\ K_5 &= \frac{\mu(12 - 6\gamma^2 - \gamma^5)}{\rho_0\gamma^{17}}, & K_6 &= \frac{\mu(12 - 4\gamma^2 - \gamma^5)}{\rho_0\gamma^{17}}, \\ K_7 &= \frac{\mu(12 - 2\gamma^2 - \gamma^5)}{\rho_0\gamma^{17}}, & K_{11} &= \frac{\mu(12 - 8\gamma^2 - \gamma^5)}{\rho_0\gamma^{17}}, \\ K_{12} &= \frac{\mu(12 - 4\gamma^2 - \gamma^5)}{\rho_0\gamma^{17}}, & K_{13} &= \frac{\mu(12 - \gamma^5)}{\rho_0\gamma^{17}}. \end{aligned} \tag{6.28}$$

By substituting (6.28) into (5.22) while taking uniform tension $\sigma_0^{ij} = \sigma_0\delta^{ij}$ and $g = 1, p_0 = 0$ into account, coefficient γ can be expressed in terms of σ_0

$$\gamma = \left(1 - \frac{\sigma_0}{\mu} \right)^{-\frac{1}{5}}. \tag{6.29}$$

Thus the equivalent free energy density is given by

$$\hat{F} = \frac{\mu}{2\rho_0} \left[\left(3 - 4\frac{\sigma_0}{\mu} \right) I_1^2 - 4 \left(1 - 2\frac{\sigma_0}{\mu} \right) I_2 \right], \tag{6.30}$$

also the elastic coefficients are derived as

$$E^{ijk\ell} = \frac{\mu}{\rho_0} \delta^{ij} \delta^{k\ell} + \frac{\mu}{\rho_0} \left(1 - 2\frac{\sigma_0}{\mu} \right) (\delta^{ik} \delta^{j\ell} + \delta^{jk} \delta^{i\ell}). \tag{6.31}$$

Finally, by substituting (6.31) into (5.24), the Cauchy stress is derived as

$$\sigma^{ij} = \sigma_0\delta^{ij} + \mu \left[\frac{\partial u_k}{\partial x^k} \delta^{ij} + \left(1 - \frac{\sigma_0}{\mu} \right) \left(\frac{\partial u_j}{\partial x^i} + \frac{\partial u_i}{\partial x^j} \right) \right]. \tag{6.32}$$

From (6.32), one finds that the initially-isothermal-uniform-extension Blatz–Ko elastomer is still isotropic. But its elastic moduli are changed, i.e.,

$$\begin{aligned} \frac{\mu'}{\mu} &= \frac{\mu - \sigma_0}{\mu}, & \frac{K'}{K} &= \frac{5\mu - 2\sigma_0}{5\mu}, \\ \frac{E'}{E} &= \frac{2(\mu - \sigma_0)(5\mu - 2\sigma_0)}{5\mu(2\mu - \sigma_0)}, & \frac{\nu'}{\nu} &= \frac{2\mu}{2\mu - \sigma_0}, \end{aligned} \tag{6.33}$$

in which $\mu, K = 5\mu/3, E = 5\mu/2$ and $\nu = 1/4$ are natural values of shear, bulk, Young moduli and Poisson ratio, respectively, while μ', K', E' and ν' are the corresponding initial-states-effected ones when $\sigma_0 \neq 0$. The relations between various coefficients and σ_0/μ are plotted in Fig. 4(a). It can be concluded that $K' < K, E' < E, \mu' < \mu$ and $\nu' > \nu$ when $\sigma_0 > 0$, but quite the opposite when $\sigma_0 < 0$. It also shows that the bulk modulus is the least affected, while the shear modulus is the most, by the given initial stress.

6.3. The Mooney–Rivlin elastomer

In the third example, we aim to derive the constitutive equations of initially isothermal sheared Mooney–Rivlin elastomers. The natural strain energy density of Mooney–Rivlin elastomer is often expressed as

$$\rho^*F = c_1(G_1 - 3) + c_2(G_2 - 3). \tag{6.34}$$

According to initially isothermal incompressible constraint $\sqrt{G} = \sqrt{g^*}$, one obtains $\rho^* = \rho_0$. Then by substituting (6.25) into (6.34), F becomes

$$F = \frac{2(c_1 + 2c_2)I_1 + 4c_2I_2}{\rho_0}. \tag{6.35}$$

Thus $K_1 = 2(c_1 + 2c_2)/\rho_0$ and $K_2 = 4c_2/\rho_0$ are the only two nonzero coefficients in (5.19). Here, $x^1 = x, x^2 = y, x^3 = z$ are still Cartesian coordinates. By substituting K_1, K_2 into (5.22), one concludes that

$$\sigma_0^{ij} = 2(c_1 + 2c_2 + c_2P_1)(\delta^{ij} + P^{ij}) - 2c_2(P^{ij} + Q^{ij}) - \rho_0\delta^{ij}, \tag{6.36}$$

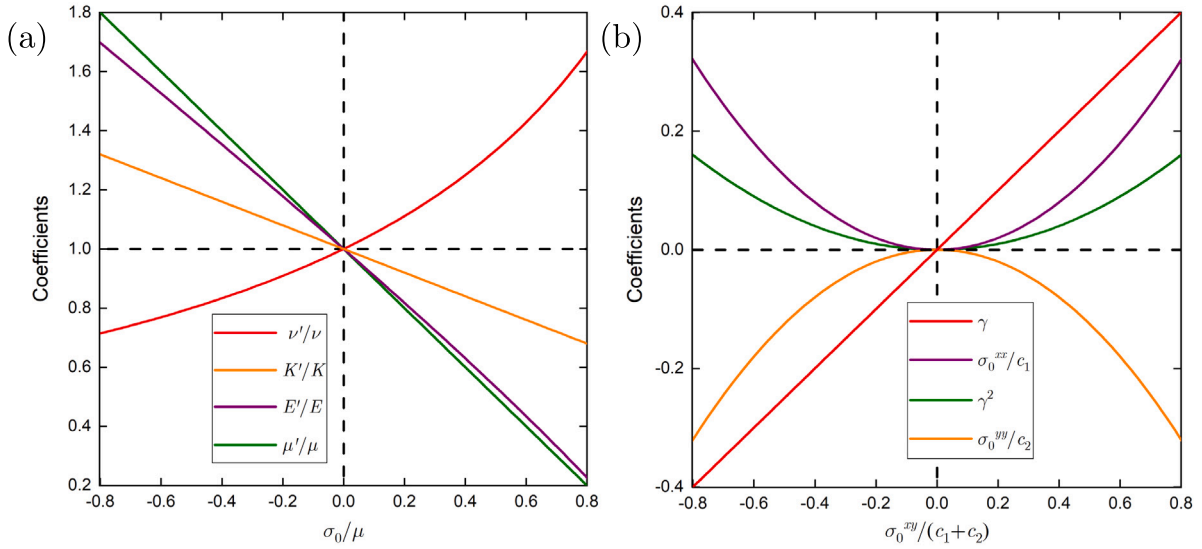


Fig. 4. Various coefficients vs dimensionless initial stresses that originated from different isothermal deformations. (a) Initially uniform tensile/compressed Blatz–Ko elastomer. (b) Initially simple sheared Mooney–Rivlin elastomer.

and the constraint equation is $\mathcal{P}_1 + \mathcal{P}_2 + \mathcal{P}_3 = 0$. To initially isothermal simple shear, the only three nonzero \mathcal{P}^{ij} are $\mathcal{P}^{xy} = \mathcal{P}^{yx} = \gamma$ and $\mathcal{P}^{xx} = \gamma^2$, thus $\mathcal{Q}^{xx} = \gamma^2 + \gamma^4, \mathcal{Q}^{xy} = \mathcal{Q}^{yx} = \gamma^3, \mathcal{Q}^{yy} = \gamma^2$ are four nonzero \mathcal{Q}^{ij} . Therefore, $\mathcal{P}_1 = -\mathcal{P}_2 = \gamma^2$ and $\mathcal{P}_3 = 0$. Note that p_0 can be determined by letting $\sigma_0^{zz} = 0$, then the solutions of algebraic equations (6.36) can be expressed as follows

$$\begin{aligned} \sigma_0^{xx} &= 2c_1\gamma^2, & \sigma_0^{yy} &= -2c_2\gamma^2, & \sigma_0^{xy} &= \sigma_0^{yx} = 2(c_1 + c_2)\gamma, \\ \sigma_0^{xz} &= \sigma_0^{zx} = \sigma_0^{yz} &= \sigma_0^{zy} &= \sigma_0^{zz} &= 0, & p_0 &= 2(c_1 + 2c_2 + c_2\gamma^2). \end{aligned} \tag{6.37}$$

Once initial shear stress σ_0^{xy} is given, γ can then be derived as $\sigma_0^{xy}/2(c_1 + c_2)$, thus all the initial stresses can be accordingly obtained. The equivalent free energy density is expressed in the form of

$$\hat{f} = \frac{p_0 I_1 + 4c_2 [(1 + \gamma^2)(I_2 - J_3 + I_1 J_1) + J_4 - I_1 J_2]}{\rho_0}. \tag{6.38}$$

We here introduce combinations $\gamma^{ij} = \mathcal{Q}^{ij} - (1 + \gamma^2)\mathcal{P}^{ij}$, which have nonzero components $\gamma^{xx} = 0, \gamma^{xy} = \gamma^{yx} = -\gamma, \gamma^{yy} = \gamma^2$. By this means, initial stresses (6.36) can be compactly expressed as $\sigma_0^{ij} = 2c_1\mathcal{P}^{ij} - 2c_2\gamma^{ij}$. According to (5.20), the elastic coefficients are expressed as the functions of γ

$$\begin{aligned} E^{ijk\ell} &= \frac{2c_2}{\rho_0} \left[(\delta^{ik}\gamma^{j\ell} + \delta^{i\ell}\gamma^{jk} + \delta^{jk}\gamma^{i\ell} + \delta^{j\ell}\gamma^{ik}) \right. \\ &\quad \left. - (1 + \gamma^2)(\delta^{ik}\delta^{j\ell} + \delta^{jk}\delta^{i\ell}) - 2\delta^{ij}\gamma^{k\ell} \right], \end{aligned} \tag{6.39}$$

where terms containing $\delta^{k\ell}$ are removed since $\delta^{k\ell}\epsilon_{k\ell} = 0$ is the linearized constraint equation. From (5.16), one concludes that $C^{ijk\ell} = -(\delta^{ik}\delta^{j\ell} + \delta^{i\ell}\delta^{jk})$ and $\chi^{ij} = \delta^{ij}$. Thus the Cauchy stresses can be derived in the following forms

$$\begin{aligned} \sigma^{ij} &= \sigma_0^{ij} + 2c_1 \left(p^{ik} \frac{\partial u_j}{\partial x^k} + p^{jk} \frac{\partial u_i}{\partial x^k} \right) + 2(c_1 + c_2) \left(\frac{\partial u_i}{\partial x^j} + \frac{\partial u_j}{\partial x^i} \right) \\ &\quad + 2c_2 \left(\gamma^{jk} \frac{\partial u_k}{\partial x^i} + \gamma^{ik} \frac{\partial u_k}{\partial x^j} \right) - 4c_2 \delta^{ij} \gamma^{k\ell} \frac{\partial u_k}{\partial x^\ell} - \delta^{ij} \hat{p}. \end{aligned} \tag{6.40}$$

Constitutive equations (6.40) behave strongly with shear-induced anisotropy. We here provide specific forms of each component of the Cauchy stresses

$$\begin{aligned}
 \sigma^{xx} &= 2c_1\gamma^2 + 4(c_1 + c_2) \left(\gamma \frac{\partial u_x}{\partial y} + \frac{\partial u_x}{\partial x} \right) + 4\gamma^2 \left(c_1 \frac{\partial u_x}{\partial x} - c_2 \frac{\partial u_y}{\partial y} \right) - \hat{p}, \\
 \sigma^{yy} &= -2c_2\gamma^2 + 4(c_1 + c_2) \left(\gamma \frac{\partial u_y}{\partial x} + \frac{\partial u_y}{\partial y} \right) - \hat{p}, \\
 \sigma^{zz} &= 4(c_1 + c_2) \frac{\partial u_z}{\partial z} + 4c_2\gamma \left(\frac{\partial u_x}{\partial y} + \frac{\partial u_y}{\partial x} - \gamma \frac{\partial u_y}{\partial y} \right) - \hat{p}, \\
 \sigma^{xy} &= 2(c_1 + c_2) \left[\gamma + \frac{\partial u_x}{\partial y} + (1 + \gamma^2) \frac{\partial u_y}{\partial x} \right] + 2(c_2 - c_1) \gamma \frac{\partial u_z}{\partial z}, \\
 \sigma^{xz} &= 2(c_1 + c_2) \left(\frac{\partial u_x}{\partial z} + \frac{\partial u_z}{\partial x} \right) + 2c_1\gamma \left(\gamma \frac{\partial u_z}{\partial x} + \frac{\partial u_z}{\partial y} \right) - 2c_2\gamma \frac{\partial u_y}{\partial z}, \\
 \sigma^{yz} &= 2(c_1 + c_2) \left(\frac{\partial u_y}{\partial z} + \frac{\partial u_z}{\partial y} \right) + 2c_2\gamma \left(\gamma \frac{\partial u_y}{\partial z} - \frac{\partial u_x}{\partial z} \right) + 2c_1\gamma \frac{\partial u_z}{\partial x}.
 \end{aligned} \tag{6.41}$$

The influences of initial stresses are reflected in coefficients γ and γ^2 . The relations between various coefficients and $\sigma_0^{xy} / (c_1 + c_2)$ are plotted in Fig. 4(b).

6.4. The Neo-Hookean elastomer

Up till now, we have derived the constitutive equations for one-dimensional unconstrained elastomers with arbitrary initial states (Example 6.1), and three-dimensional unconstrained and constrained elastomers with isothermal initial stresses (Example 6.2, 6.3). In this example, we will consider the most general situation, i.e., to derive the thermo-mechanically coupled constitutive equations for Neo-Hookean elastomers with arbitrary initial states.

The natural strain energy density of Neo-Hookean elastomer is expressed as

$$\rho^* \mathcal{F} = kT (\mathcal{G}_1 - 3), \tag{6.42}$$

in which k is the material parameter concerning the Boltzmann constant and the chain density. Here, we employ temperature-controlled compressible constraint (5.12), where $\zeta(T) = \xi(T - T^*)$, and coordinates x^i are still Cartesian, thus $\rho^* = \rho_0 / (1 - \xi\vartheta^*)$. Then one obtains the following free energy density

$$\mathcal{F} = \frac{2k(1 - \xi\vartheta^*)(T_0 + \vartheta) I_1}{\rho_0}. \tag{6.43}$$

By substituting (6.43) into (5.19), the only three nonzero coefficients are

$$K_1 = \frac{2kT_0}{\rho_0} (1 - \xi\vartheta^*), \quad K_4 = \frac{k\mathcal{P}_1}{\rho_0} (1 - \xi\vartheta^*), \quad K_8 = \frac{2k(1 - \xi\vartheta^*)}{\rho_0}. \tag{6.44}$$

Then by substituting (6.44) into (5.22), algebraic equations take the forms of

$$\frac{\sigma_0^{ij}}{kT_0} + \frac{p_0\delta^{ij}}{kT_0} = 2(1 - \xi\vartheta^*) (\delta^{ij} + \mathcal{P}^{ij}), \quad 1 + \mathcal{P}_1 + \mathcal{P}_2 + \mathcal{P}_3 = (1 - \xi\vartheta^*)^{-2}. \tag{6.45}$$

Also from (3.4) and (4.27), the initial and the reference entropies are derived as

$$s_0 = \frac{6kT_0(1 - \xi\vartheta^*) - (3 + 2\xi T_0)p_0 - \mathcal{L}_1}{2\rho_0 T_0}, \quad s^* = -\frac{2k\xi T^*(1 - \xi\vartheta^*)}{\rho_0}. \tag{6.46}$$

In summary, the equivalent free energy density can be expressed as

$$\hat{\mathcal{F}} = \frac{p_0(I_1 - \xi\vartheta) + 2k(1 - \xi\vartheta^*)(I_1 + \mathcal{J}_1)\vartheta}{\rho_0}, \tag{6.47}$$

while according to (5.20), (5.21) and (6.45), one obtains $E^{ijk\ell} = 0$, and

$$\psi^{ij} = -\frac{\sigma_0^{ij}}{\rho_0 T_0} - \frac{p_0\delta^{ij}}{\rho_0 T_0}. \tag{6.48}$$

Thus the Cauchy stresses and the entropy can be derived as

$$\begin{aligned} \sigma^{ij} &= (1 - \xi\vartheta) \sigma_0^{ij} + p_0 \left(\frac{\partial u_j}{\partial x^i} + \frac{\partial u_i}{\partial x^j} - 2\xi\vartheta\delta^{ij} \right) + \sigma_0^{jk} \frac{\partial u_i}{\partial x^k} + \sigma_0^{ik} \frac{\partial u_j}{\partial x^k} \\ &\quad - (1 + \xi\vartheta) \hat{p} \delta^{ij} + \left(\sigma_0^{ij} + p_0 \delta^{ij} \right) \frac{\vartheta}{T_0}, \\ s &= \frac{3k(1 - \xi\vartheta^*)}{\rho_0} - \frac{p_0 + \mathcal{L}_1}{2\rho_0 T_0} - \frac{\sigma_0^{ij}}{\rho_0 T_0} \frac{\partial u_i}{\partial x^j} - \frac{(1 + \xi\vartheta) [p_0 + \xi T_0 (p_0 + \hat{p})]}{\rho_0 T_0}, \\ 1 &= \frac{p_0^3 + S_1 p_0^2 + S_2 p_0 + S_3}{(1 - \xi\vartheta^*) (2kT_0)^3}, \end{aligned} \tag{6.49}$$

where $S_1 = \mathcal{L}_1$, $S_2 = \frac{1}{2} (\mathcal{L}_1^2 - \mathcal{L}_2)$, $S_3 = \frac{1}{6} (\mathcal{L}_1^3 - 3\mathcal{L}_1\mathcal{L}_2 + 2\mathcal{L}_3)$ are three principal invariants of σ_0^{ij} . (6.49)₃ is the thermo-mechanically coupled version of the determining equation of initial constraint multiplier p_0 . If $\xi = 0$, there is no thermal effect and (6.49)₃ degenerates to the classical form of Gower, Ciarletta, and Destrade (2015), i.e., $p_0^3 + S_1 p_0^2 + S_2 p_0 + S_3 = (2kT_0)^3$. Once σ_0^{ij} and ϑ^* are given, one can solve cubic equation (6.49)₃ to obtain $p_0 = p_0(S_1, S_2, S_3, \vartheta^*)$, then by substituting it into (6.45), $\mathcal{P}^{ij} = \mathcal{P}^{ij}(\sigma_0^{ij}, \vartheta^*)$ can be accordingly obtained. Thus the thermo-mechanically coupled constitutive equations of Neo-Hookean elastomers with arbitrary initial states are derived.

The simplest situation of the initial states is that they are originated from initially isothermal uniform tensions/compressions, thus $\vartheta^* = 0$ and $\sigma_0^{ij} = \sigma_0 \delta^{ij}$. Due to the incompressible constraint, there is no initial deformation, i.e., $\mathcal{P}^{ij} = 0$. Since $S_1 = 3\sigma_0$, $S_2 = 3\sigma_0^2$, $S_3 = \sigma_0^3$, then (6.49)₃ and (6.46)₁ are solved as

$$\frac{p_0}{2kT_0} = 1 - \frac{\sigma_0}{2kT_0}, \quad \frac{\rho_0 s_0}{k} = 2\xi T_0 \left(\frac{\sigma_0}{2kT_0} - 1 \right). \tag{6.50}$$

Equivalent free energy density (6.47) now becomes

$$\hat{F} = \frac{(2kT_0 - \sigma_0) (\mathcal{I}_1 - \xi\vartheta) + 2k\mathcal{I}_1\vartheta}{\rho_0}. \tag{6.51}$$

And constitutive equations (6.49) take the following forms

$$\begin{aligned} \sigma^{ij} &= 2kT_0 \left(\frac{\partial u_i}{\partial x^j} + \frac{\partial u_j}{\partial x^i} - 2\xi\vartheta\delta^{ij} \right) + [2k\vartheta + (1 + \xi\vartheta) (\sigma_0 - \hat{p})] \delta^{ij}, \\ s &= \frac{2k}{\rho_0} [1 - (1 + \xi T_0) (1 + \xi\vartheta)] + \frac{\xi(1 + \xi\vartheta)}{\rho_0} (\sigma_0 - \hat{p}). \end{aligned} \tag{6.52}$$

If one regards $\sigma_0 - \hat{p}$ as a new constraint multiplier, the constitutive equations remain unchanged. This fact indicates that initially isothermal uniform tensions/compressions nearly do not influence the subsequent thermal and mechanical performances of Neo-Hookean elastomers.

In the second situation, the initial states are assumed to be originated from initially isothermal uniaxial tensions/compressions along the z direction, where σ_0^{zz} is the only nonzero initial stress, thus $S_1 = \sigma_0^{zz}$, $S_2 = S_3 = 0$. We here introduce coefficient $\gamma = \sqrt{1 + \mathcal{P}^{zz}}$, then $\mathcal{P}^{zz} = \gamma^2 - 1$ and $\mathcal{P}^{xx} = \mathcal{P}^{yy} = \gamma^{-1} - 1$. By this means, cubic equation (6.49)₃ and initial entropy (6.46)₁ are solved as

$$\frac{\sigma_0^{zz}}{2kT_0} = \gamma^2 - \frac{1}{\gamma}, \quad \frac{p_0}{2kT_0} = \frac{1}{\gamma}, \quad \frac{\rho_0 s_0}{k} = 3 - \gamma^2 - \frac{2(1 + \xi T_0)}{\gamma}. \tag{6.53}$$

Equivalent free energy density (6.47) now becomes

$$\hat{F} = \frac{2kT_0 (\mathcal{I}_1 - \xi\vartheta) + 2k [\mathcal{I}_1 + (\gamma^3 - 1) \varepsilon_{zz}] \vartheta}{\gamma \rho_0}. \tag{6.54}$$

And constitutive equations (6.49) take the following forms

$$\begin{aligned} \sigma^{ij} &= \frac{2kT_0}{\gamma} \left(\frac{\partial u_j}{\partial x^i} + \frac{\partial u_i}{\partial x^j} - 2\xi\vartheta\delta^{ij} \right) + \left[\frac{2k\vartheta}{\gamma} - (1 + \xi\vartheta) \hat{p} \right] \delta^{ij} \\ &\quad + \sigma_0^{zz} \left[\delta^{iz} \frac{\partial u_j}{\partial z} + \delta^{jz} \frac{\partial u_i}{\partial z} + \left(1 + \frac{\vartheta}{T_0} - \xi\vartheta \right) \delta^{iz} \delta^{jz} \right], \\ s &= \frac{2k}{\rho_0 \gamma} \left[\frac{3 - \gamma^2}{2\gamma^{-1}} + (1 - \gamma^3) \frac{\partial u_z}{\partial z} - (1 + \xi T_0) (1 + \xi\vartheta) \right] - \frac{\xi(1 + \xi\vartheta)}{\rho_0} \hat{p}. \end{aligned} \tag{6.55}$$

According to (6.55), one concludes that the influences of initially isothermal uniaxial tensions/compressions on the constitutive equations are mainly reflected in coefficients γ , γ^2 and γ^3 , and the initial-states-effected material parameter is $k' = k/\gamma$. The relations between various coefficients and σ_0^{zz}/kT_0 are plotted in Fig. 5(a). It can be concluded that both γ , γ^2 and γ^3 increase with σ_0^{zz} , while $k' < k$ when $\sigma_0^{zz} > 0$, but quite the opposite when $\sigma_0^{zz} < 0$. More importantly, $\rho_0 s_0/k' \leq \rho^* s^*/k$, that the initial entropy is always kept decreasing. If $\xi = 0$, by taking the γ derivative of (6.53)₃, one obtains

$$-\rho_0 T_0 \frac{\partial s_0}{\partial \gamma} = \frac{2kT_0}{\gamma} \left(\gamma^2 - \frac{1}{\gamma} \right) = \frac{\sigma_0^{zz}}{\gamma}. \tag{6.56}$$

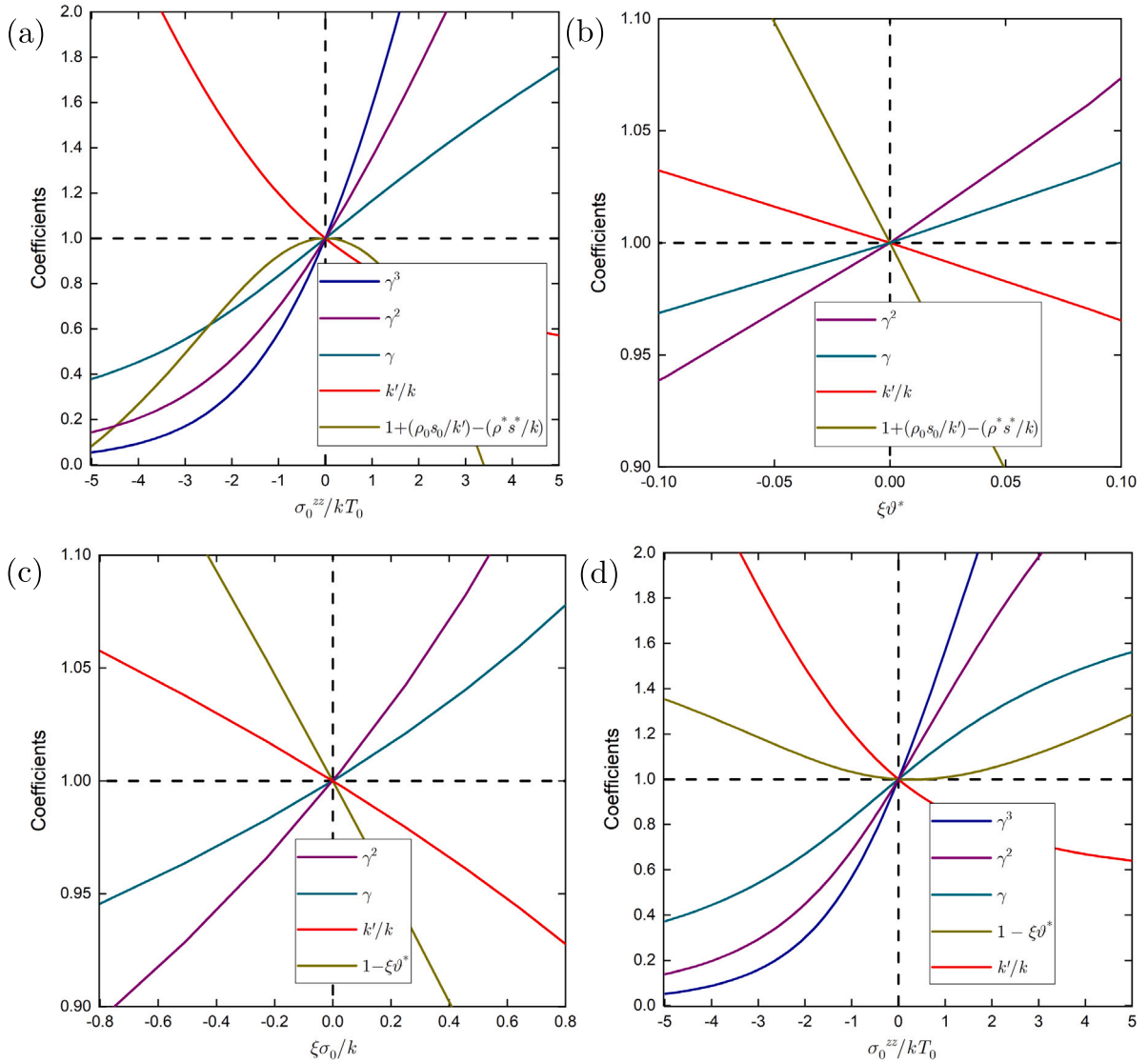


Fig. 5. Various coefficients vs dimensionless initial states that originated from different thermo-mechanically coupled deformation histories. (a) Initially isothermal uniaxial tension/compression (b) Initially free thermal expansion (c) Initially adiabatic uniform tension/compression (d) Initially adiabatic uniaxial tension/compression.

(6.56) is the classical formula for describing isothermally uniaxial tension in rubber-like entropy elasticity. In addition, according to (6.53)₁, one can specify $\Sigma(\gamma_t) = 2kT_0(\gamma_t^2 - \gamma_t^{-1})$. Then from (6.22), the multiplicative-decomposition-based axial Cauchy stress can be expressed in the form of

$$\sigma = \frac{2kT_0}{\gamma_0} [\gamma_0^3 - 1 + (2\gamma_0^3 + 1) \epsilon_e]. \tag{6.57}$$

By substituting $\vartheta = 0, \partial u_x / \partial x = \partial u_y / \partial y = -\frac{1}{2} \partial u_z / \partial z = -\frac{1}{2} \epsilon_e$ and $\sigma^{xx} = \sigma^{yy} = 0$ into (6.55)₁, σ^{zz} is equal to σ only and only if $\gamma = \gamma_0$. This verifies once more the equivalence of the two approaches in isothermal situations.

In the third situation, the initial states are assumed to be originated from initially free thermal expansions, where $\sigma_0^{ij} = 0$ and $\mathcal{P}^{ij} = \mathcal{P} \delta^{ij}$, thus $S_1 = S_2 = S_3 = 0$. We here introduce coefficient $\gamma = \sqrt{1 + \mathcal{P}}$, then it can be derived that

$$\frac{p_0}{2kT_0} = \frac{1}{\gamma}, \quad 1 - \xi \vartheta^* = \frac{1}{\gamma^3}, \quad \frac{\rho_0 s_0}{k} = \frac{3(1 - \gamma^2) - 2\xi T_0 \gamma^2}{\gamma^3}. \tag{6.58}$$

Equivalent free energy density (6.47) now becomes

$$\hat{F} = \frac{2kT_0 (I_1 - \xi\vartheta) + 2kI_1\vartheta}{\gamma\rho_0}. \tag{6.59}$$

And constitutive equations (6.49) take the following forms

$$\begin{aligned} \sigma^{ij} &= \frac{2kT_0}{\gamma} \left(\frac{\partial u_i}{\partial x^j} + \frac{\partial u_j}{\partial x^i} - 2\xi\vartheta\delta^{ij} \right) + \left[\frac{2k\vartheta}{\gamma} - (1 + \xi\vartheta)\hat{p} \right] \delta^{ij}, \\ s &= \frac{2k}{\rho_0\gamma} \left[\frac{3 - \gamma^2}{2\gamma^2} - (1 + \xi T_0)(1 + \xi\vartheta) \right] - \frac{\xi(1 + \xi\vartheta)}{\rho_0} \hat{p}. \end{aligned} \tag{6.60}$$

According to (6.60), the influences of initially free thermal expansions are mainly reflected in coefficients γ and γ^2 , and the initial-states-effected material parameter is also $k' = k/\gamma$. The relations between various coefficients and $\xi\vartheta^*$ are plotted in Fig. 5(b). It can be concluded that γ and γ^2 increase with ϑ^* , while $k' < k, \rho_0 s_0/k' < \rho^* s^*/k$ when $\vartheta^* > 0$, but quite the opposite when $\vartheta^* < 0$.

In the fourth situation, the initial states are assumed to be originated from initially adiabatic uniform tensions/compressions, where $\sigma_0^{ij} = \sigma_0\delta^{ij}, \mathcal{P}^{ij} = \mathcal{P}\delta^{ij}$ and $s_0 = s^*$. We here still employ coefficient $\gamma = \sqrt{1 + \mathcal{P}}$. Then according to (6.46), cubic equation (6.49)₃ can be solved as

$$\begin{aligned} \frac{\xi\sigma_0}{k} &= \frac{\gamma^3(3 + 2\xi T_0)(\gamma^2 - 1) + 2(\gamma^3 - 1)}{\gamma^6}, \quad 1 - \xi\vartheta^* = \frac{1}{\gamma^3}, \\ \frac{\rho_0}{2kT_0} &= \frac{1}{\gamma} - \frac{\sigma_0}{2kT_0} = \frac{\gamma^3 - 3\gamma^5 + 2 + 2\gamma^3\xi T_0}{2\gamma^6\xi T_0}. \end{aligned} \tag{6.61}$$

Similar to (6.11), σ_0 and ϑ^* are not independent. Suppose σ_0 is given and ϑ^* can be accordingly determined. By this means, (6.47) takes the following forms

$$\hat{F} = \frac{(2kT_0 - \gamma\sigma_0)(I_1 - \xi\vartheta) + 2kI_1\vartheta}{\gamma\rho_0}. \tag{6.62}$$

And constitutive equations (6.49) take the following forms

$$\begin{aligned} \sigma^{ij} &= \frac{2kT_0}{\gamma} \left(\frac{\partial u_i}{\partial x^j} + \frac{\partial u_j}{\partial x^i} - 2\xi\vartheta\delta^{ij} \right) + \left[\frac{2k\vartheta}{\gamma} + (1 + \xi\vartheta)(\sigma_0 - \hat{p}) \right] \delta^{ij}, \\ s &= \frac{2k}{\rho_0\gamma} \left[\frac{3 - \gamma^2}{2\gamma^2} - (1 + \xi T_0)(1 + \xi\vartheta) \right] + \frac{\xi(1 + \xi\vartheta)}{\rho_0} (\sigma_0 - \hat{p}). \end{aligned} \tag{6.63}$$

The influences of initially adiabatic uniform tensions/compressions are also reflected in coefficients γ and γ^2 , while the initial-states-effected material parameter and constraint multiplier are $k' = k/\gamma, \sigma_0 - \hat{p}$, respectively. In Humphrey and Rajagopal (1997), the authors suggested that $\xi = 5 \times 10^{-4} \text{ K}^{-1}$. If T^* is assumed to be 300 K, then $\xi T_0 = 1.15 - \gamma^{-3}$. The relations between various coefficients and $\xi\sigma_0/k$ are plotted in Fig. 5(c). It can be concluded that both γ and γ^2 increase with σ_0 , while $k' < k, \vartheta^* > 0$ when $\sigma_0 > 0$, but quite the opposite when $\sigma_0 < 0$. If $\xi = 0$, then $\gamma = 1$ and $\sigma_0 = 0$, which means that there is no adiabatic uniform tension/compression for incompressible constraint.

In the last situation, the initial states are assumed to be originated from initially adiabatic uniaxial tensions/compressions along the z direction, where $s_0 = s^*$, while σ_0^{zz} is the only nonzero initial stress. Here, we still employ coefficient $\gamma = \sqrt{1 + \mathcal{P}^{zz}}$, then according to (6.46), one concludes that

$$\begin{aligned} \frac{\sigma_0^{zz}}{2kT_0} &= \left(1 - \frac{1}{\gamma} \right) \frac{\gamma(\gamma + 1)(5 + 2\xi T^*) + 2}{2 - \gamma^3 + \gamma(3 + 2\xi T^*)}, \quad \frac{\rho_0}{2kT_0} = \frac{1}{\gamma}, \\ \mathcal{P}^{xx} = \mathcal{P}^{yy} &= \frac{2 - \gamma^3 - \gamma}{2\gamma(2 + \xi T^*)}, \quad 1 - \xi\vartheta^* = \frac{2(2 + \xi T^*)}{2 - \gamma^3 + \gamma(3 + 2\xi T^*)}. \end{aligned} \tag{6.64}$$

Equivalent free energy density (6.47) now can be expressed in the form of

$$\hat{F} = \frac{2kT_0 (I_1 - \xi\vartheta) + (\gamma\sigma_0^{zz}\varepsilon_{zz} + 2kT_0 I_1) (\vartheta/T_0)}{\gamma\rho_0}. \tag{6.65}$$

And constitutive equations (6.49) take the following forms

$$\begin{aligned} \sigma^{ij} &= \frac{2kT_0}{\gamma} \left(\frac{\partial u_i}{\partial x^j} + \frac{\partial u_j}{\partial x^i} - 2\xi\vartheta\delta^{ij} \right) + \left[\frac{2k\vartheta}{\gamma} - (1 + \xi\vartheta)\hat{p} \right] \delta^{ij} \\ &\quad + \sigma_0^{zz} \left[\delta^{iz} \frac{\partial u_j}{\partial z} + \delta^{jz} \frac{\partial u_i}{\partial z} + \left(1 + \frac{\vartheta}{T_0} - \xi\vartheta \right) \delta^{iz} \delta^{jz} \right], \\ s &= \frac{2k}{\rho_0\gamma} \left[\frac{\gamma(3 - \gamma^2)(2 + \xi T^*)}{2 - \gamma^3 + \gamma(3 + 2\xi T^*)} - (1 + \xi\vartheta)(1 + \xi T_0) \right. \\ &\quad \left. - \frac{\gamma(\gamma^2 - 1)(5 + 2\xi T^*) + 2(\gamma - 1) \frac{\partial u_z}{\partial z}}{2 - \gamma^3 + \gamma(3 + 2\xi T^*)} \right] - \frac{\xi(1 + \xi\vartheta)}{\rho_0} \hat{p}. \end{aligned} \tag{6.66}$$

The influences of initially adiabatic uniaxial tensions/compressions are mainly reflected in coefficient γ , γ^2 , and γ^3 , and the initial-states-effected material parameter is also $k' = k/\gamma$. Here, $\xi T^* = 0.15$ is also employed. The relations between various coefficients and σ_0^{zz}/kT_0 are plotted in Fig. 5(d). It can be concluded that γ , γ^2 and γ^3 increase with σ_0 , while $k' < k$ when $\sigma_0 > 0$, but quite the opposite when $\sigma_0 < 0$. More importantly, $\vartheta^* \leq 0$, that the initial temperature is always kept decreasing. If $\xi = 0$, then $\gamma = 1$ and $\sigma_0^{zz} = 0$, there are also no adiabatic uniaxial tensions/compressions for incompressible constraint.

Compare constitutive equations (6.55) and (6.55), corresponding to isothermal and adiabatic situations, respectively, not only significant differences can be found between these formulations, but also different relationships between γ and σ_0^{zz} . By further comparing Figs. 5(a) and 5(c), it also shows that the thermal coefficients are the most influenced by the thermo-mechanically coupled deformation histories, while the mechanical coefficients behave similarly and only small quantitative differences can be found. It demonstrated once again that the constitutive coefficients may behave differently for the same initial stresses that are originated from distinct deformation histories.

To illustrate the rationality of the change of thermal response with tensile or compressive prestress, we here quote the experiment of Joule et al. (1859) and the simulation of Lev, Faye, and Volokh (2019). We here use heat capacity as an example. According to the natural state-based material model of Lev et al. (2019), a purely thermal term should be added to the free energy density (6.43), i.e.,

$$F = \frac{2k(T_0 + \vartheta) I_1}{\rho_0} + c_v T^* \ln\left(\frac{T_0 + \vartheta}{T^*}\right), \tag{6.67}$$

where incompressible constraint is employed ($\xi = 0$ and $\rho^* = \rho_0$) and c_v is the natural heat capacity. By substituting (6.67) into (5.19), one obtains

$$K_1 = \frac{2kT_0}{\rho_0}, \quad K_4 = \frac{kP_1}{\rho_0} + \frac{c_v T^*}{T_0}, \quad K_8 = \frac{2k}{\rho_0}, \quad K_{14} = -\frac{c_v T^*}{T_0^2}. \tag{6.68}$$

Then by substituting (6.68) into (5.22), (3.4) and (5.23), the initial and the reference entropies can be derived as

$$s_0 = -\frac{kP_1}{\rho_0} - \frac{c_v T^*}{T_0}, \quad s^* = -c_v. \tag{6.69}$$

Here, the initial states are assumed to be originated from initially adiabatic uniaxial tensions along the z direction, where $s_0 = s^*$, while σ_0^{zz} is the only nonzero initial stress. By introducing coefficient $\gamma = \sqrt{1 + P^{zz}}$, one obtains

$$\frac{\sigma_0^{zz}}{2kT_0} = \gamma^2 - \frac{1}{\gamma}, \quad \frac{\vartheta^*}{T^*} = \frac{\bar{k}(\gamma^3 - 3\gamma + 2)}{\gamma - \bar{k}(\gamma^3 - 3\gamma + 2)}, \tag{6.70}$$

where $\bar{k} = k/(\rho_0 c_v)$ is a dimensionless material constant. According to the Lev et al. (2019), the reference heat capacity can be expressed in the form $c'_v = c_v(T^*/T_0)$. Then, according to (6.70), the following formula can be derived

$$\frac{c'_v}{c_v} = 1 - \frac{\bar{k}(\gamma^3 - 3\gamma + 2)}{\gamma}. \tag{6.71}$$

Compared with the experimental data of Joule et al. (1859), we find the best fit curve corresponds to a small material constant $\bar{k} = 3.9 \times 10^{-4}$. Fig. 6a illustrates that our prediction of the initial temperature increment is close to the experiment results of Joule et al. (1859) and also the simulation of Lev et al. (2019). It can be concluded that initial temperature increases with initial stretch. Then, according to the expression $c'_v = c_v(T^*/T_0)$, reference heat capacity is the function of the initial temperature. Thus, the presence of initially adiabatic tensions can indeed affect the reference heat capacity (Fig. 6b). However, for some real materials, \bar{k} is very small, and hence the relative variation of heat capacity is slight. Consequently, thermal responses are often regarded as independent of the initial stress.

Finally, by substituting $\sigma_0 = \sigma_0^{zz} = 0$ and $\gamma = 1$ into (6.52), (6.55), (6.60), (6.63) and (6.66), all these constitutive equations degenerate to

$$\begin{aligned} \sigma^{ij} &= 2kT_0 \left(\frac{\partial u_i}{\partial x^j} + \frac{\partial u_j}{\partial x^i} - 2\xi\vartheta\delta^{ij} \right) + [2k\vartheta - (1 + \xi\vartheta)\hat{p}]\delta^{ij}, \\ s &= \frac{2k}{\rho_0} [1 - (1 + \xi T_0)(1 + \xi\vartheta)] - \frac{\xi(1 + \xi\vartheta)}{\rho_0}\hat{p}. \end{aligned} \tag{6.72}$$

Further assume that $\xi = 0$, (6.72) reduce to the following classical forms

$$\sigma^{ij} = 2kT_0 \left(\frac{\partial u_i}{\partial x^j} + \frac{\partial u_j}{\partial x^i} \right) + (2k\vartheta - \hat{p})\delta^{ij}, \quad s = 0. \tag{6.73}$$

6.5. The Gent elastomer and the exponential form elastomer

In the last example, we aim to derive constitutive equations for residually stressed thin-wall tube made of Gent elastomer and exponential form elastomer, where cylindrical material coordinates $x^1 = r, x^2 = \varphi, x^3 = z$ are employed. Thus $g_{rr} = g^{rr} = 1, g_{\varphi\varphi} = r^2, g^{\varphi\varphi} = r^{-2}, g_{zz} = g^{zz} = 1, g_{r\varphi} = g_{r\varphi} = g_{\varphi z} = g^{r\varphi} = g^{r\varphi} = g^{\varphi z} = 0$ and $\sqrt{g} = r$. The tube has internal and external radii r_i and r_o , while wall thickness $r_o - r_i$ is much smaller than r_i . The residual stresses are assumed to be originated from initially isothermal deformations.

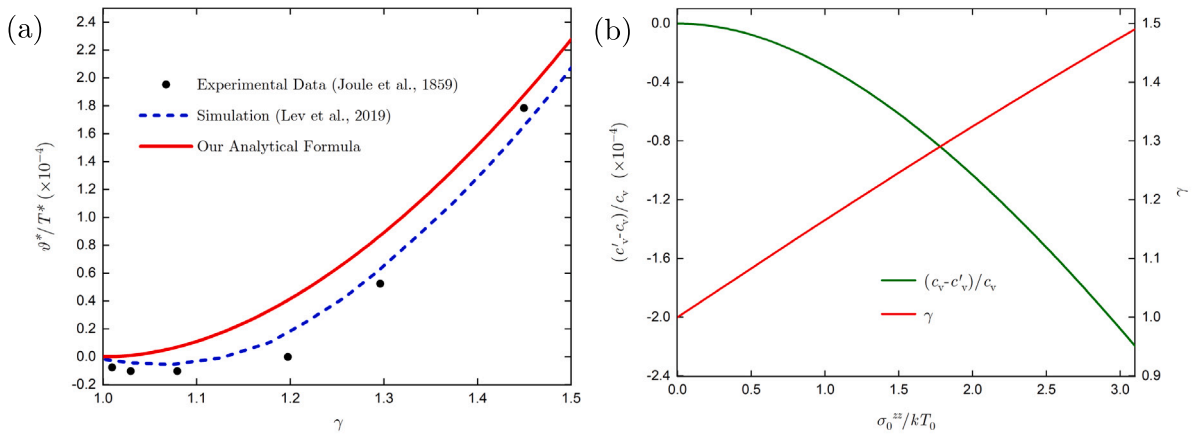


Fig. 6. (a) Initial temperature increment versus stretch for $\bar{k} = 3.9 \times 10^{-4}$. (b) The change of heat capacity with tensile prestress (which originated from initial adiabatic stretch).

The natural strain energy density of Gent elastomer is often expressed as

$$\rho^* F = -\frac{\mu\lambda}{2} \ln\left(1 - \frac{\mathcal{G}_1 - 3}{\lambda}\right), \tag{6.74}$$

where μ, λ are material parameters. Suppose $\xi = 0$, then one obtains

$$F = -\frac{\mu\lambda}{2\rho_0} \ln\left(1 - \frac{2I_1}{\lambda}\right). \tag{6.75}$$

By substituting (6.75) into (5.19), the only two nonzero coefficients are

$$K_1 = \frac{\mu\lambda}{\rho_0(\lambda - \mathcal{P}_1)}, \quad K_{11} = \frac{2\mu\lambda}{\rho_0(\lambda - \mathcal{P}_1)^2}. \tag{6.76}$$

Then according to (5.22), the algebraic equations can be expressed as

$$\sigma_0^{ij} + rp_0 g^{ij} = \frac{\mu\lambda}{\lambda - \mathcal{P}_1} (g^{ij} + \mathcal{P}^{ij}). \tag{6.77}$$

Similar to (6.49)₃, the Gent version of the determining equation of p_0 is

$$\begin{aligned} & [(\lambda + 3)^3 - 27] (rp_0)^3 + [(\lambda + 3)^3 S_1 - 27(S_1 + \mu\lambda)] (rp_0)^2 \\ & + [(\lambda + 3)^3 S_2 - 9(S_1 + \mu\lambda)^2] (rp_0) = (S_1 + \mu\lambda)^3 - (\lambda + 3)^3 S_3. \end{aligned} \tag{6.78}$$

Once S_1, S_2, S_3 are given, one can solve cube Eq. (6.78) to derive $p_0 = p_0(S_1, S_2, S_3)$, then by substituting it into (6.77), $\mathcal{P}^{ij} = \mathcal{P}^{ij}(S_1, S_2, S_3)$ can be accordingly obtained. When $\lambda \rightarrow \infty$, Gent elastomer degenerates to Neo-Hookean elastomer, and (6.78) reduces to $(rp_0)^3 + S_1(rp_0)^2 + S_2(rp_0) + S_3 = 1$.

Similarly, the natural strain energy density of exponential form elastomer is

$$\rho^* F = \frac{\mu\lambda}{2} \left(e^{\frac{\mathcal{G}_1 - 3}{\lambda}} - 1 \right), \tag{6.79}$$

where μ, λ are still material parameters. Thus the free energy density becomes

$$F = \frac{\mu\lambda}{2\rho_0} e^{\frac{2I_1}{\lambda}}. \tag{6.80}$$

By substituting (6.79) into (5.19), the only two nonzero coefficients are

$$K_1 = \frac{\mu}{\rho_0} e^{\frac{\mathcal{P}_1}{\lambda}}, \quad K_{11} = \frac{2\mu}{\rho_0\lambda} e^{\frac{\mathcal{P}_1}{\lambda}}. \tag{6.81}$$

Then according to (5.22), the algebraic equations can be expressed as

$$\sigma_0^{ij} + rp_0 g^{ij} = \mu e^{\frac{\mathcal{P}_1}{\lambda}} (g^{ij} + \mathcal{P}^{ij}). \tag{6.82}$$

Different from (6.78), here \mathcal{P}_1 but not p_0 , is taken as the unknown variable, i.e.,

$$\frac{\mu^3 (27 + 9\mathcal{P}_1 + \mathcal{P}_1^2) \mathcal{P}_1 e^{\frac{3\mathcal{P}_1}{\lambda}} + 3\mu (3S_2 - S_1^2) (3 + \mathcal{P}_1) e^{\frac{\mathcal{P}_1}{\lambda}}}{9S_1 S_2 - 27S_3 - 2S_1^3} = 1. \tag{6.83}$$

Transcendental equation (6.83) is the exponential version of the determining equation of p_0 . Once S_1, S_2, S_3 are given, one can solve (6.83) to derive $\mathcal{P}_1 = \mathcal{P}_1(S_1, S_2, S_3)$, then $p_0 = p_0(S_1, S_2, S_3)$ can be correspondingly determined by substituting it into $3rp_0 = \mu \left(3 + \mathcal{P}_1\right) e^{\frac{p_1}{\lambda}} - S_1$. After that, $\mathcal{P}^{ij} = \mathcal{P}^{ij}(S_1, S_2, S_3)$ can be derived from (6.82). When $\lambda \rightarrow \infty$, exponential form elastomer also degenerates to Neo-Hookean elastomer, and (6.83) reduces to $\mu^3 (27 + 9\mathcal{P}_1 + \mathcal{P}_1^2) \mathcal{P}_1 + 3\mu (3S_2 - S_1^2) (3 + \mathcal{P}_1) = 9S_1S_2 - 27S_3 - 2S_1^3$, which is the same as classical form $(rp_0)^3 + S_1 (rp_0)^2 + S_2 (rp_0) + S_3 = 1$ when the rp_0 is substituted by $\mu \left(1 + \frac{p_1}{3}\right) - \frac{S_1}{3}$. In this respect, the two types of elastomers can be regarded as the generalizations of Neo-Hookean elastomers.

In Merodio and Ogden (2016), σ_0^{ij} are specified in quadratic forms

$$\begin{aligned} \sigma_0^{rr} &= \frac{\kappa\mu}{r_i^2} \left[r^2 - (r_o + r_i) r + r_o r_i \right], & \sigma_0^{zz} &= 0, \\ r^2 \sigma_0^{\varphi\varphi} &= \frac{\kappa\mu}{r_i^2} \left[3r^2 - 2 (r_o + r_i) r + r_o r_i \right], \end{aligned} \tag{6.84}$$

while in Du et al. (2018), σ_0^{ij} are specified in logarithmic forms

$$\begin{aligned} \sigma_0^{rr} &= \kappa\mu \ln \left(\frac{r}{r_i} \right) \ln \left(\frac{r}{r_o} \right), & \sigma_0^{zz} &= 0, \\ r^2 \sigma_0^{\varphi\varphi} &= \kappa\mu \left[\ln \left(\frac{r}{r_i} \right) + \ln \left(\frac{r}{r_o} \right) + \ln \left(\frac{r}{r_i} \right) \ln \left(\frac{r}{r_o} \right) \right], \end{aligned} \tag{6.85}$$

where κ is the parameter concerning the intensity of the given residual stresses. We here introduce the following dimensionless variable h and parameter H

$$h = \frac{r}{r_i} - 1, \quad H = \frac{r_o}{r_i} - 1, \quad 0 \leq h \leq H \ll 1. \tag{6.86}$$

By substituting (6.86) into (6.84) and (6.85), one concludes that both Gent elastomer and exponential form elastomer correspond to the same linearization

$$\sigma_0^{rr} = 0, \quad r^2 \sigma_0^{\varphi\varphi} = \kappa\mu (2h - H). \tag{6.87}$$

Then according to (6.83), the same linearized solutions can also be obtained

$$\begin{aligned} rp_0 &= \mu \left[1 + \frac{\kappa(H - 2h)}{3} \right], & \mathcal{P}_1 &= 0, \\ p^{rr} = p^{zz} &= \frac{\kappa(H - 2h)}{3}, & r^2 p^{\varphi\varphi} &= \frac{2\kappa(2h - H)}{3}. \end{aligned} \tag{6.88}$$

By this means, the two types of elastomers have the same constitutive equations. The subsequent deformations are specified in the following forms

$$u^r = \frac{a}{r_i(1+h)}, \quad u^\varphi = u^z = 0, \quad \nabla_r u^r = -\nabla_\varphi u^\varphi = -\frac{a}{r_i^2(1+h)^2}, \tag{6.89}$$

where a is an undetermined coefficient. Then by substituting (6.76)₂ and (6.84) into (5.24), as well as taking the equilibrium equations and boundary conditions $\sigma^{rr}|_{h=0} = -P, \sigma^{rr}|_{h=H} = 0$ into account, the Cauchy stresses can be derived as

$$\begin{aligned} \sigma^{rr} &= \frac{h-H}{H} P, & \sigma^{zz} &= \frac{2-H}{4H} P + \frac{\kappa(H-2h)P}{6H}, \\ r^2 \sigma^{\varphi\varphi} &= \frac{2-2h+H}{2H} P + \kappa(2h-H) \left(\mu + \frac{P}{6H} \right), \end{aligned} \tag{6.90}$$

in which P is the internal pressure of the thin-wall tube. In comparison with the existing results of the implicit constitutive theory of Bustamante and Rajagopal (2018), we specify $H = 0.1, \mu/P = 500$ and $\kappa = 0, 0.04, 0.08, 0.12, 0.16, 0.2$ for different intensities of the residual stresses. The distributions of principal stresses for different intensities of the residual stresses are plotted in Fig. 7. Our linearized solutions show a good qualitative agreement with the implicit constitutive theory, especially in the center of the tube. When $\kappa = 0$, both results reduce to the classical solutions of linear elasticity.

7. Conclusions

This work establishes a general framework for deriving constitutive equations for soft elastomers with arbitrary initial states. Instead of using the virtual stress-free configuration, we define the *natural state* by imposing the stress-free condition and the natural temperature condition. The derivations are based on the new proposed *intrinsic embedding method of initial states*, in which an additive decomposition of material strains is employed while the influences of different thermo-mechanically coupled deformation histories are taken into account. In this way can the material coordinates be properly defined. Once the natural-state-based free energy density and internal constraint are specified, the required constitutive equations can be accordingly obtained.

Despite its inherently greater complexity, the intrinsic embedding method of initial states is appropriate for describing initially-stressed elastomers in multiple coupled fields. The full incorporation of thermal effects is the example. Here, what matters is the additive decomposition of strain. If the initial stain is supposed to be originated from previous elastic-plastic, electromagnetic-elastic, or strain-gradient-elasticity-based deformations, new models concerning various effects can be conveniently established

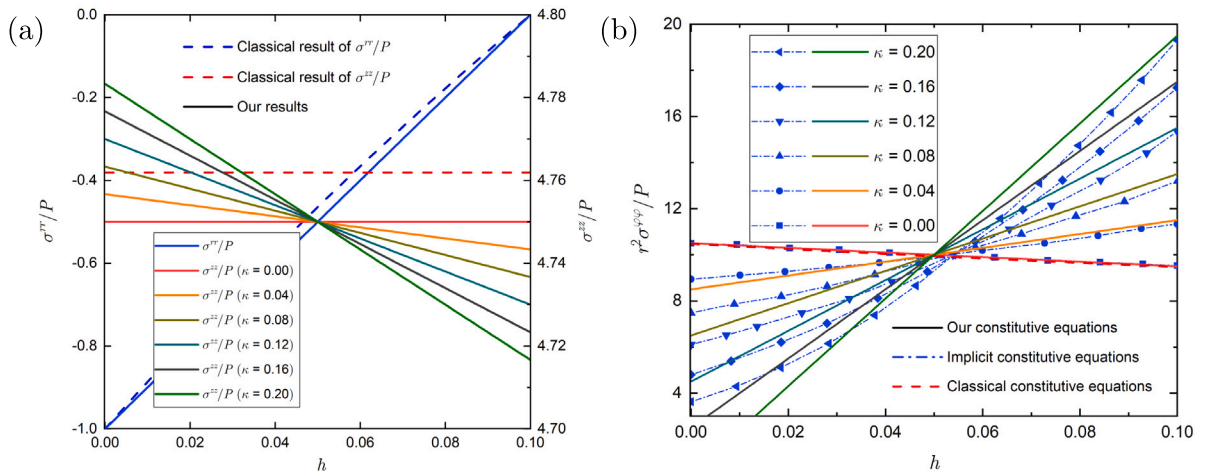


Fig. 7. Distributions of principal stresses for different intensities of the residual stresses. (a) Radial and axial stresses (b) Circumferential stress.

based on the proposed embedding method. Moreover, as a modular approach, we have provided many readily available formulae for isotropic elastomers. Engineers can use them directly according to the actual situations. Further investigations of the generalization of this theory to anisotropic situations are underway.

The explicit formulations of the Cauchy stress and the entropy are derived from the linearizations of the obtained constitutive equations. Then we embed the initial states in hyperelastic models of Saint Venant–Kirchhoff, Blatz–Ko, Mooney–Rivlin, Neo-Hookean, Gent, and exponential form. The influences brought by the initial stresses that originated from various isothermal or adiabatic deformations, the initial temperature produced by uniformly thermal expansions, and the temperature-controlled internal constraint on the elastic coefficients are analyzed, respectively. Compared with the classical and the implicit constitutive theories, the new proposed constitutive equations not only show quantitative agreement with the above two under isothermal circumstances but also present some novel properties concerning different thermo-mechanically coupled conditions. These results perfect the constitutive theory of soft materials and can be further developed by applying them to engineering practices.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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