



# On the two-potential constitutive modeling of rubber viscoelastic materials



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## ABSTRACT

This Note lays out the specialization of the two-potential constitutive framework – also known as the “generalized standard materials” framework – to rubber viscoelasticity. *Inter alia*, it is shown that a number of popular rubber viscoelasticity formulations, introduced over the years following different approaches, are special cases of this framework. As a first application of practical relevance, the framework is utilized to put forth a new objective and thermodynamically consistent rubber viscoelastic model for incompressible isotropic elastomers. The model accounts for the non-Gaussian elasticity of elastomers, as well as for the deformation-enhanced shear thinning of their viscous dissipation governed by reptation dynamics. The descriptive and predictive capabilities of the model are illustrated via comparisons with experimental data available from the literature for two commercially significant elastomers.

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## 1. Introduction

While within certain ranges of loading conditions elastomers may be safely idealized as elastic, they are intrinsically viscoelastic. In the literature, there are, in essence, two approaches that have been adopted to model their viscoelastic behavior at a continuum level. The first approach is grounded on hereditary integral representations of the stress (strain) in terms of the strain (stress) (see, e.g., [1–3]), whereas the second one is based on the employment of internal variables to describe their viscous dissipation (see, e.g., [4–9]). Because of its superior tractability, the internal-variables approach has proved overwhelmingly more popular. Now, within the theoretical description of dissipative (not just viscoelastic) phenomena based on internal variables [10], the so-called two-potential framework [11–14] – also known as the “generalized standard materials” framework – has emerged as a framework of choice to construct thermodynamically consistent models for a wide range of phenomena such as for instance plasticity [15], fracture [16], and ferroelectricity [17]. Yet, somewhat surprisingly, the specialization of this framework to the case of rubber viscoelasticity does not appear to have been reported in the literature.<sup>1</sup>

The purpose of this Note is to lay out the two-potential constitutive framework for rubber viscoelasticity. The consistency of the framework with material frame indifference, material symmetry, and the second law of thermodynamics is placed on record. As an illustration of the generality of the framework, it is also shown that a number of popular rubber viscoelasticity formulations, which have been introduced over the years using different approaches, can all be derived from the two-potential constitutive framework.

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<sup>1</sup> There are, however, a number of models in the literature that have been proposed using directly this approach (see, e.g., [18,19]).

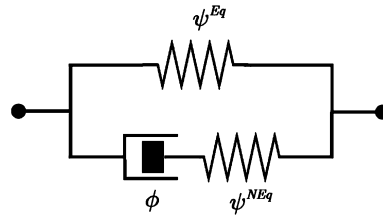


Fig. 1. Rheological model of rubber viscoelastic behavior.

A second objective of this Note is to put forth, by means of the two-potential framework, a new objective and thermodynamically consistent viscoelastic model for the practically relevant case of isotropic incompressible elastomers. The model accounts for the non-Gaussian elasticity of elastomers, as well as for the deformation-enhanced shear thinning of their viscous dissipation governed by the reptational motion of the underlying polymer chains. The computational tractability and robustness of the model is illustrated together with its predictive and descriptive capabilities via comparisons with experimental data available from the literature for two commercially significant elastomers, Nitrile rubber and the acrylate elastomer VHB 4910 from 3M.

## 2. The two-potential framework for rubber viscoelasticity

*The two-potential framework in mechanics* The key idea behind the two-potential constitutive framework is to describe the manner in which a material stores and dissipates energy by means of two thermodynamic potentials: (i) a free energy function  $\psi$  and (ii) a dissipation potential  $\phi$ . In the context of mechanics [11–14], formally,

$$\psi = \psi(\mathbf{F}, \mathbf{\Lambda}) \quad \text{and} \quad \phi = \phi(\mathbf{F}, \mathbf{\Lambda}, \dot{\mathbf{\Lambda}}) \tag{1}$$

under isothermal conditions, where  $\mathbf{F}$  is the deformation gradient tensor,  $\mathbf{\Lambda}$  stands for a finite number of (macroscopically non-observable) internal variables, and  $\dot{\mathbf{\Lambda}} = d\mathbf{\Lambda}/dt$  denotes the time derivative of  $\mathbf{\Lambda}$ . Both of these potentials are required to be non-negative and objective functions. In addition, the dissipation potential  $\phi$  is required to be convex in  $\dot{\mathbf{\Lambda}}$  and such that  $\text{argmin}_{\dot{\mathbf{\Lambda}}} \phi(\mathbf{F}, \mathbf{\Lambda}, \dot{\mathbf{\Lambda}}) = \mathbf{0}$ . The constitutive relation implied by these potentials is given by the following two coupled equations:

$$\mathbf{S} = \frac{\partial \psi}{\partial \mathbf{F}}(\mathbf{F}, \mathbf{\Lambda}), \quad \frac{\partial \psi}{\partial \mathbf{\Lambda}}(\mathbf{F}, \mathbf{\Lambda}) + \frac{\partial \phi}{\partial \dot{\mathbf{\Lambda}}}(\mathbf{F}, \mathbf{\Lambda}, \dot{\mathbf{\Lambda}}) = \mathbf{0} \tag{2}$$

where  $\mathbf{S}$  stands for the first Piola–Kirchhoff stress tensor and the differential equation in time (2)<sub>2</sub> is commonly referred to as the evolution equation for the internal variables  $\mathbf{\Lambda}$ . By virtue of the objectivity of  $\psi$  and  $\phi$  and the convexity of  $\phi$  in  $\dot{\mathbf{\Lambda}}$ , the constitutive relation (2) satisfies the principle of material frame indifference and the second law of thermodynamics. Moreover, it satisfies automatically the balance of angular momentum  $\mathbf{S}\mathbf{F}^T = \mathbf{F}\mathbf{S}^T$ .

*Rubber viscoelasticity* The specialization of the above constitutive framework to rubber viscoelasticity, much like to any other type of dissipative phenomenon, amounts to selecting appropriate internal variables  $\mathbf{\Lambda}$  and appropriate thermodynamic potentials  $\psi$  and  $\phi$ . For rubber viscoelasticity, this selection must account for at least five features (two microscopic and three macroscopic in nature) that have been well established experimentally about elastomers: (i) the storage of energy is primarily governed by changes in entropy of the underlying polymer network, (ii) the dissipation of energy is primarily governed by friction among neighboring polymer chains, (iii) when all forces are removed after an arbitrary loading path, elastomers creep to their original configuration, (iv) when subjected to relaxation and creep loading conditions, elastomers exhibit a transient response that then evolves into an equilibrium state of deformation and stress, and (v) when subjected to loading conditions of the same type but different loading rate, elastomers exhibit different responses.

In the classical context of small-deformation linear viscoelastic behavior, features (iii) through (v) are encompassed by the basic rheological model depicted in Fig. 1. This model suggests that the internal variables  $\mathbf{\Lambda}$  should be identified, loosely speaking, with a deformation gradient,  $\mathbf{F}^v$  say, associated with the viscous (i.e. dissipative) part of the deformation, and moreover that

$$\psi = \psi^{\text{Eq}}(\mathbf{F}) + \psi^{\text{NEq}}(\mathbf{F}, \mathbf{F}^v) \quad \text{and} \quad \phi = \phi(\mathbf{F}, \mathbf{F}^v, \dot{\mathbf{F}}^v) \tag{3}$$

where the free energy function  $\psi^{\text{Eq}}$  serves to characterize the thermodynamic equilibrium states of the elastomer, while  $\psi^{\text{NEq}}$  serves to account for the additional energy storage at non-equilibrium states.

To proceed further, it is necessary to establish the precise definition of  $\mathbf{F}^v$  and how the non-equilibrium part  $\psi^{\text{NEq}}$  of the free energy depends on it. Consistent with the rheological model depicted in Fig. 1 and motivated by earlier efforts [4], we take  $\chi$ , the mapping between material points  $\mathbf{X}$  in the undeformed reference configuration  $\Omega_0$  and their spatial position  $\mathbf{x}$  in the deformed configuration  $\Omega$ , to be given by the composition

$$\chi(\mathbf{X}) = \chi_e \circ \chi_v = \chi_e(\chi_v(\mathbf{X})) = \chi_e(\xi) \quad \text{with} \quad \xi = \chi_v(\mathbf{X}) \tag{4}$$

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