

Constitutive Model for Rubberlike Materials

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Abstract

The isotropic continuum stored energy (CSE) functional, fully combined with the Poisson function, has been applied to constitutively model compressible as well as incompressible rubberlike materials. The isotropic CSE constitutive model fits uniaxial tension test and predicts unfitted pure shear and equibiaxial tension tests of incompressible S4035A thermoplastic elastomer (TPE). Furthermore, the isotropic CSE model along with the Poisson function fits uniaxial tension test along with the kinematic relation test and predicts unfitted pure shear and equibiaxial tension tests along with the corresponding kinematic relation tests of a compressible synthetic rubber. The detailed procedures for uniquely identifying constitutive parameters are emphasized, the experimental characterization results are discussed, and the principles for constitutive models are summarized and augmented.

Keywords

Compressible Finite Elasticity, Constitutive Modeling, Isotropic CSE Functional, Poisson Function, Rubberlike Materials

1. Introduction

Natural rubbers and synthetic elastomers with a wide range of applications, including but not limited to tires, engine mounts, seals, dampers, hoses, tunnel linings, and bump stoppers, can be classified as isotropic hyperelastic materials. Relevant isotropic properties are inherited from long polymeric chains with a high degree of flexibility and mobility. Rubbers can be dense with little cavities like many fluids and they are nearly incompressible. On the other hand, all materials are compressible to a certain degree so that rubberlike materials are generally compressible. Sulfur vulcanization is a fundamental manufacturing process that converts natural rubbers and synthetic elastomers into crosslinked rubbers and elastomers, achieving desired properties of materials and perfor-

mances of products. Rubberlike materials, as an isotropic hyperelastic material, exhibit a nonlinear finite elastic deformation capability. For constitutive modeling of isotropic hyperelastic materials such as rubbers and elastomers, the existence of the Helmholtz free-energy function, Ψ , was a classical postulation. When hyperelastic materials are idealized as homogeneous materials, the Helmholtz free-energy function depends only on a finite deformation measure and Ψ is referred to as the stored energy function per unit reference volume as documented by Holzapfel (2000) [1]. Thus, the stored energy density function can be constructed as a continuous and scalar-valued function of the right Cauchy-Green tensor \mathbf{C}

$$\Psi = \Psi(\mathbf{C}), \tag{1}$$

in which $\mathbf{C} = \mathbf{F}^T \mathbf{F}$, and \mathbf{F} is the deformation gradient tensor.

For perfectly hyperelastic materials, no entropy is locally produced. The internal dissipation is zero $\mathcal{D}_{\text{int}} = 0$. The constitutive equation can be directly derived from the Clausius-Planck form of the second law of thermodynamics. With $\dot{\Psi} = \partial\Psi(\mathbf{C})/\partial\mathbf{C} : \dot{\mathbf{C}}$, the zero internal dissipation gives

$$\mathcal{D}_{\text{int}} = \mathbf{S} : \frac{\dot{\mathbf{C}}}{2} - \dot{\Psi} = \left[\mathbf{S} - 2 \frac{\partial\Psi(\mathbf{C})}{\partial\mathbf{C}} \right] : \frac{\dot{\mathbf{C}}}{2} = 0, \tag{2}$$

where \mathbf{C} and $\dot{\mathbf{C}}$ are arbitrary, the expression in the square bracket must be zero which yields a relatively general constitutive equation for isotropic hyperelastic materials

$$\mathbf{S} = 2 \frac{\partial\Psi(\mathbf{C})}{\partial\mathbf{C}}, \tag{3}$$

where \mathbf{S} is the second Piola-Kirchhoff stress tensor.

For isotropic hyperelastic materials, the stored energy density function can be expressed uniquely in terms of the three scalar invariants of the right Cauchy-Green tensor, which was established by the classical work of Rivlin (1948) [2].

$$\Psi = \Psi[I_1(\mathbf{C}), I_2(\mathbf{C}), I_3(\mathbf{C})], \tag{4}$$

where the three principal scalar invariants of the right Cauchy-Green tensor are defined by

$$I_1(\mathbf{C}) = \text{tr}\mathbf{C}, \quad I_2(\mathbf{C}) = \frac{1}{2}[(\text{tr}\mathbf{C})^2 - \text{tr}\mathbf{C}^2], \quad I_3(\mathbf{C}) = \det\mathbf{C}. \tag{5}$$

Taking derivatives with the chain rule by Substituting (4) into (3), the general constitutive equation reads

$$\mathbf{S} = 2 \left(\frac{\partial\Psi}{\partial I_1} \frac{\partial I_1}{\partial\mathbf{C}} + \frac{\partial\Psi}{\partial I_2} \frac{\partial I_2}{\partial\mathbf{C}} + \frac{\partial\Psi}{\partial I_3} \frac{\partial I_3}{\partial\mathbf{C}} \right), \tag{6}$$

where the derivatives of the three invariants with respect to \mathbf{C} read

$$\frac{\partial I_1}{\partial\mathbf{C}} = \mathbf{I}, \quad \frac{\partial I_2}{\partial\mathbf{C}} = I_1 \mathbf{I} - \mathbf{C}, \quad \frac{\partial I_3}{\partial\mathbf{C}} = I_3 \mathbf{C}^{-1}. \tag{7}$$

Substituting (7) into (6) gives the most general form of a stress relation in terms of the three stretch-based invariants, which model isotropic rubberlike materials at finite deformations as

$$\mathbf{S} = 2 \left[\left(\frac{\partial \Psi}{\partial I_1} + I_1 \frac{\partial \Psi}{\partial I_2} \right) \mathbf{I} - \frac{\partial \Psi}{\partial I_2} \mathbf{C} + I_3 \frac{\partial \Psi}{\partial I_3} \mathbf{C}^{-1} \right], \quad (8)$$

where the stored energy density function, Ψ , needs to be determined. The continuum stored energy function was coined since Equation (8) was established based on continuum mechanics.

Mechanical responses of rubberlike materials under loadings are commonly characterized by isotropic nonlinear stress-stretch relationships. Mechanical behaviors of rubberlike materials can be captured with the constitutive model formulated through the mathematical concept of symmetry, the physical principle of energy balance, experimental evidence of multiple deformation modes, and kinematic relationships. The effectiveness of constitutive models plays a key role in the predictive capabilities of computational models for structures in finite element analyses.

Perfect incompressibility is assumed for rubberlike materials, making analytical solutions of problems with simple geometry easier, but numerical finite element implementations more difficult. As the Poisson's ratio gets closer to 0.5 for perfect incompressibility, the numerical accuracy, stability, and efficiency of finite element models deteriorate. Thus, the physical modeling demands compressible formulations as emphasized by Fried and Johnson (1988) [3]. To model the compressibility, bulk moduli are traditionally used, establishing the pressure-volume relationships. The stress and deformation states in uniaxial tension, pure shear, and equibiaxial tension modes as examples, however, can never match the pressure and volume states. Furthermore, when volumes are used, more detailed geometric dimensions are missing. For compressible rubberlike materials, more detailed kinematic relationships are needed in experimental characterizations. In order to model the actual stress and deformation states for compressible rubberlike materials, the isotropic CSE functional will be fully combined with the Poisson function. Many experimental results on kinematic relationships follow the Poisson functions, in which Poisson's ratio is defined in terms of true strains or natural logarithms of stretches. The Poisson functions in uniaxial tension, pure shear, and equibiaxial tension modes can be found in the article by Blatz and Ko (1962) [4].

The major objectives, therefore, are to fully combine the isotropic CSE functional with the Poisson function to constitutively model and predict finite elastic deformations of compressible as well as incompressible rubberlike materials. Experimental characterization requirements in uniaxial tension, pure shear, and equibiaxial tension modes are briefly described. The detailed methods and procedures for identifying constitutive parameters are summarized. The principles of constitutive models are summarized and augmented.

This paper is organized as follows. In Section 2, some thoughts on the formu-

lation of the isotropic CSE functional are summarized, the isotropic CSE functional is fully combined with the Poisson functions to formulate constitutive equations in uniaxial tension, pure shear, and equibiaxial tension modes, and experimental characterization requirements are briefly described. In Section 3, uniquely identifying constitutive parameters are emphasized, described, and applied to fit uniaxial tension test data for incompressible S4035A TPE and compressible synthetic rubber. The prediction checks in pure shear and equibiaxial tension modes are also emphasized for both cases. In Section 4, both the experimental characterization results and the principles for constitutive models are discussed. In Section 5, conclusions are drawn.

2. Nonlinear Finite Elasticity

2.1. Isotropic CSE Functional

Mathematical models of natural laws are frequently formulated through the concept of symmetry into nonlinear partial differential equations.

For constitutive modeling of isotropic rubberlike materials with nonlinear finite elastic deformations, the second Piola-Kirchhoff stress tensor (8), \mathbf{S} , has been derived. For work conjugate pairs, a finite deformation measure is needed. The commonly used finite deformation measures are the right Cauchy-Green tensor \mathbf{C} , the Green-Lagrange strain tensor \mathbf{E} , and the deformation gradient tensor \mathbf{F} . The right Cauchy-Green tensor is a deformation tensor that excludes rigid body rotation in \mathbf{F} . Furthermore, experimental stress-stretch curves of isotropic rubberlike materials are strictly increasing functions, which are injective. Additionally, every stress on the curves must correspond to at least one stretch, which requires a surjective function. A function that is both injective and surjective is also called a bijective function in terms of abstract algebra. For constructing a bijective function, the stretch-based tensor \mathbf{C} rather than the strain tensor \mathbf{E} is more appropriately selected since stretches are always represented by positive real numbers. More specifically, the strain tensor \mathbf{E} could create singularities and imaginary numbers in constitutive modelings. Thus, the second Piola-Kirchhoff stress tensor \mathbf{S} and the right Cauchy-Green tensor \mathbf{C} as a work conjugate pair have been selected to construct the isotropic CSE functional.

For a physically consistent, mathematically covariant, and geometrically meaningful formulation, the isotropic CSE functional was postulated and balanced with its stress work done based on the conservation of energy by Zhao (2016) [5]

$$\Psi = \mathbf{S} : \frac{\mathbf{C}}{2}. \quad (9)$$

The isotropic CSE functional (9) is covariant to $\Psi_E = \mathbf{S} : \mathbf{E}$ under the transformation of $\mathbf{E} = 0.5(\mathbf{C} - \mathbf{I})$. Substituting (8) into (9), simplifying, and rearranging yields the partial differential equation of the isotropic CSE functional in terms of three invariants I_1 , I_2 , and I_3

$$\Psi = I_1 \frac{\partial \Psi}{\partial I_1} + 2I_2 \frac{\partial \Psi}{\partial I_2} + 3I_3 \frac{\partial \Psi}{\partial I_3}. \quad (10)$$

With Lie group methods, the characteristic system of the partial differential Equation (10) takes the form of

$$\frac{dI_1}{I_1} = \frac{dI_2}{2I_2} = \frac{dI_3}{3I_3} = \frac{d\Psi}{\Psi}. \quad (11)$$

Taking its independent first-integrals, $\psi_1 = I_2/I_1^2$, $\psi_2 = I_3/I_1^3$, and $\psi_3 = \Psi/I_1$, the general solution to the isotropic CSE partial differential Equation (10) has been obtained as

$$\Psi = I_1 h(I_2/I_1^2, I_3/I_1^3), \quad (12)$$

where h as an arbitrary function has been selected as the summation of two arbitrary functions of invariant groups albeit other available combinations

$$\Psi = I_1 \left[f(I_2/I_1^2) + g(I_3/I_1^3) \right], \quad (13)$$

where f and g as two arbitrary functions that can be determined by curvatures of deformations. For normal and shear deformations, the first arbitrary function, f , has been defined as

$$f(I_2/I_1^2) = c_1 + c_2 \sqrt{I_2/I_1^2} = c_1 + c_2 \frac{\sqrt{I_2}}{I_1}, \quad (14)$$

and for different degrees of ellipsoidal deformations, the second arbitrary function, g , has been generalized as

$$g(I_3/I_1^3) = c_3 (I_3/I_1^3)^{-c_4} = c_3 \frac{I_1^{3c_4}}{I_3^{c_4}}. \quad (15)$$

Substituting (14) and (15) into (13) augments the isotropic CSE functional by Zhao (2020) [6]. Applying the normalization condition, $\Psi(I) = 0$, gives

$$\Psi = c_1 (I_1 - 3) + c_2 (\sqrt{I_2} - \sqrt{3}) + c_3 \left(\frac{I_1^{3c_4+1}}{I_3^{c_4}} - 3^{3c_4+1} \right), \quad (16)$$

where the four constitutive parameters, c_1 , c_2 , c_3 , and c_4 , will be identified by experimental tests. Thus, the isotropic CSE functional will be used to establish constitutive equations of commonly used deformation modes in experimental tests.

2.2. Isotropic CSE Constitutive Equations

Nominal stress and stretch results are preferably calculated from force and extension measurements with original sample dimensions recorded in experimental tests. The first Piola-Kirchhoff stress tensor or the nominal stress tensor \mathbf{P} is related to the second Piola-Kirchhoff stress tensor \mathbf{S} through the deformation gradient tensor \mathbf{F} by $\mathbf{P} = \mathbf{F}\mathbf{S}$ or more conveniently by

$$\mathbf{P}^T = \frac{\partial \Psi}{\partial \mathbf{F}} = 2 \left(\frac{\partial \Psi}{\partial I_1} \frac{\partial I_1}{\partial \mathbf{C}} + \frac{\partial \Psi}{\partial I_2} \frac{\partial I_2}{\partial \mathbf{C}} + \frac{\partial \Psi}{\partial I_3} \frac{\partial I_3}{\partial \mathbf{C}} \right) \left(\frac{1}{2} \frac{\partial \mathbf{C}}{\partial \mathbf{F}} \right) = \mathbf{S}\mathbf{F}^T, \quad (17)$$

where the nominal stresses as a function of stretches in indicial notation are

generally expressed as

$$P_{ji} = \frac{\partial \Psi}{\partial I_1} \frac{\partial I_1}{\partial \lambda_{ij}} + \frac{\partial \Psi}{\partial I_2} \frac{\partial I_2}{\partial \lambda_{ij}} + \frac{\partial \Psi}{\partial I_3} \frac{\partial I_3}{\partial \lambda_{ij}}, \quad (i, j = 1, 2, 3). \tag{18}$$

The three derivatives of the isotropic CSE functional (16) with respect to invariants I_1 , I_2 , and I_3 are worked out as

$$\frac{\partial \Psi}{\partial I_1} = c_1 + c_3(3c_4 + 1) \frac{I_1^{3c_4}}{I_3^{c_4}}, \quad \frac{\partial \Psi}{\partial I_2} = \frac{c_2}{2\sqrt{I_2}}, \quad \frac{\partial \Psi}{\partial I_3} = -c_3c_4 \frac{I_1^{3c_4+1}}{I_3^{c_4+1}}. \tag{19}$$

Thus, the isotropic CSE constitutive equations will be specifically derived from Equations (16) through (19), combining with the Poisson functions based on stretch and stress states at different deformation modes.

Experimental characterizations of rubberlike materials are often conducted by uniaxial tension, uniaxial compression, equibiaxial tension, biaxial tension, pure shear, and simple shear tests. Among them, uniaxial tension, pure shear, and equibiaxial tension tests are the most commonly used testing methods for finite element analyses as reviewed by Charlton *et al.* (1994) [7]. The experimental characterization requirements along with the constitutive equations are described in the uniaxial tension, pure shear, and equibiaxial tension modes.

2.3. Uniaxial Tension Mode

In a uniaxial tension (UT) test, the stretch state is $\lambda_1 = \lambda = L/L_0$ and $\lambda_2 = \lambda_3 = \lambda^{-\nu}$ based on the UT Poisson function where the current effective length L and original effective length L_0 must be measured on the specimen away from the clamps by a video extensometer or laser extensometer, respectively. Lateral contractions λ_2 and λ_3 should also be measured as a function of uniaxial stretch λ_1 for compressible materials. The stress state is preferably calculated by nominal stress as $P_1 = P/A_0$ and $P_2 = P_3 = 0$ where the applied load P is measured by a load cell and the original cross-section area A_0 can be measured by a caliper. The specimen with the aspect ratio of length to width or thickness should be greater than 10 in order to reduce lateral constraint to thinning. For uniform clamping, a dumbbell-shaped specimen is often used. The three invariants in the uniaxial tension mode are obtained from the stretch state as

$$I_1^{ut} = \lambda^2 + 2\lambda^{-2\nu}, \quad I_2^{ut} = 2\lambda^{2-2\nu} + \lambda^{-4\nu}, \quad I_3^{ut} = \lambda^{2-4\nu}. \tag{20}$$

The nominal stress as a function of principal stretch in uniaxial tension mode for rubberlike materials, along with the stress-free condition, can be generally derived as

$$\begin{aligned} P_{ut} = & 2 \left[\lambda - 2\nu\lambda^{-2\nu-1} - (1-2\nu) \right] c_1 + 2 \left[\frac{(1-\nu)\lambda^{1-2\nu} - \nu\lambda^{-4\nu-1}}{\sqrt{2\lambda^{2-2\nu} + \lambda^{-4\nu}}} - \frac{1-2\nu}{\sqrt{3}} \right] c_2 \\ & + 2 \left\{ \frac{(\lambda^2 + 2\lambda^{-2\nu})^{3c_4}}{\lambda^{(2-4\nu)c_4}} \left[(3c_4 + 1)(\lambda - 2\nu\lambda^{-2\nu-1}) \right. \right. \\ & \left. \left. - c_4(1-2\nu)(\lambda + 2\lambda^{-2\nu-1}) \right] - (1-2\nu)27^{c_4} \right\} c_3. \end{aligned} \tag{21}$$

For the assumed incompressible condition, the equation (21) with the Poisson's ratio $\nu = 0.5$ is simplified as

$$\tilde{P}_{ur} = 2(\lambda - \lambda^{-2})c_1 + \frac{1 - \lambda^{-3}}{\sqrt{2\lambda + \lambda^{-2}}}c_2 + 2(3c_4 + 1)(\lambda^2 + 2\lambda^{-1})^{3c_4}(\lambda - \lambda^{-2})c_3. \quad (22)$$

2.4. Pure Shear or Planar Tension Mode

In a pure shear (PS) or planar tension test, the stretch state is $\lambda_1 = \lambda = L/L_0$, $\lambda_2 = 1$, and $\lambda_3 = \lambda^{-\nu/(1-\nu)}$ with the PS Poisson function where the current and original effective lengths must be measured on the specimen away from the clamps by a video extensometer or laser extensometer, respectively. Thickness reduction λ_3 should also be measured as a function of axial stretch λ_1 for compressible materials. The stress state is preferably calculated by nominal stress as $P_1 = P/A_0$, $P_2 \neq 0$, and $P_3 = 0$ where the applied load P is measured by a load cell and the original cross-section area can be measured by a caliper. The specimen with the aspect ratio of width to length should be at least 10 in order to constrain the width and let all specimen thinning occur in the thickness direction. Because the material is nearly incompressible, a state of pure shear exists in the specimen at a 45-degree angle to the stretching direction. The three invariants in the pure shear mode are obtained from the stretch state as

$$I_1^{ps} = \lambda^2 + 1 + \lambda^{\frac{2\nu}{1-\nu}}, \quad I_2^{ps} = \lambda^2 + \lambda^{\frac{2-4\nu}{1-\nu}} + \lambda^{\frac{2\nu}{1-\nu}}, \quad I_3^{ps} = \lambda^{\frac{2-4\nu}{1-\nu}}. \quad (23)$$

The nominal stress as a function of principal stretch in pure shear mode for rubberlike materials, along with the stress-free condition, can be generally worked out as

$$\begin{aligned} P_{ps} = & 2 \left(\lambda - \frac{\nu}{1-\nu} \lambda^{\frac{1+\nu}{1-\nu}} - \frac{1-2\nu}{1-\nu} \right) c_1 \\ & + \left[\frac{\lambda - \frac{\nu}{1-\nu} \lambda^{\frac{1+\nu}{1-\nu}} + \frac{1-2\nu}{1-\nu} \lambda^{\frac{1-3\nu}{1-\nu}}}{\sqrt{\lambda^2 + \lambda^{\frac{2-4\nu}{1-\nu}} + \lambda^{\frac{2\nu}{1-\nu}}}} - \left(\frac{1-2\nu}{1-\nu} \right) \frac{2}{\sqrt{3}} \right] c_2 \\ & + 2 \left\{ \frac{\left(\lambda^2 + 1 + \lambda^{\frac{2\nu}{1-\nu}} \right)^{3c_4}}{\lambda^{\frac{2-4\nu}{1-\nu}c_4}} \left[(3c_4 + 1) \left(\lambda - \frac{\nu}{1-\nu} \lambda^{\frac{1+\nu}{1-\nu}} \right) \right. \right. \\ & \left. \left. - c_4 \frac{1-2\nu}{1-\nu} \left(\lambda + \lambda^{-1} + \lambda^{\frac{1+\nu}{1-\nu}} \right) \right] - \frac{1-2\nu}{1-\nu} 27^{c_4} \right\} c_3. \end{aligned} \quad (24)$$

For the assumed incompressible condition, $\nu = 0.5$, Equation (24) is simplified as

$$\tilde{P}_{ps} = 2(\lambda - \lambda^{-3})c_1 + \frac{\lambda - \lambda^{-3}}{\sqrt{\lambda^2 + \lambda^{-2} + 1}}c_2 + 2(3c_4 + 1)(\lambda^2 + \lambda^{-2} + 1)^{3c_4}(\lambda - \lambda^{-3})c_3. \quad (25)$$

2.5. Equibiaxial Tension Mode

The biaxial tension (BT) test involves one of the most complicated deformations among the available tests because of the number of independent variable stretches. In general, five variables, λ_1 , λ_2 , λ_3 , κ_1 , and κ_2 , have to be measured in the deformation gradient tensor

$$\mathbf{F}_{bt} = \begin{pmatrix} \lambda_1 & \kappa_1 & 0 \\ \kappa_2 & \lambda_2 & 0 \\ 0 & 0 & \lambda_3 \end{pmatrix}, \tag{26}$$

When constant ratios of λ_1 to λ_2 are used, the number of independent variables can be reduced, especially for equibiaxial tension tests $\lambda_1 = \lambda_2$. In practical applications, however, shear stretches are usually ignored albeit shear stretches are generally two orders of magnitude smaller than normal stretches, i.e. $\kappa \approx 10^{-2} \lambda$ as studied by Aydin *et al.* (2017) [8].

In an equibiaxial tension (ET) test, the simplified stretch state is $\lambda_1 = \lambda_2 = \lambda = L/L_0$ and $\lambda_3 = \lambda^{-2\nu/(1-\nu)}$ based on the ET Poisson function. The current effective length L and original effective length L_0 must be measured on the specimen away from the clamps by a video extensometer or laser extensometer, similarly. Thickness reduction λ_3 should also be measured as a function of equibiaxial stretches λ_1 and λ_2 for compressible materials. The stress state is $P_1 = P_2 = P/A_0$ and $P_3 = 0$ where the applied load P is measured by a load cell and the original cross-section area A_0 can be measured by a caliper. Stress-stretch curves in equibiaxial tension tests of square specimens could be overestimated since parts of applied loads are also used to generate shear stretches rather than just normal stretches. The stress-stretch curves could be underestimated due to branching stability issues from a bifurcation point of view.

The three invariants in equibiaxial tension mode are obtained from the simplified stretch state as

$$I_1^{et} = 2\lambda^2 + \lambda^{\frac{4\nu}{1-\nu}}, \quad I_2^{et} = \lambda^4 + 2\lambda^{\frac{2-6\nu}{1-\nu}}, \quad I_3^{et} = \lambda^{\frac{4-8\nu}{1-\nu}}. \tag{27}$$

The nominal stress as a function of principal stretch in equibiaxial tension mode for rubberlike materials, along with the stress-free condition, turns out to be

$$P_{et} = 2 \left(\lambda - \frac{\nu}{1-\nu} \lambda^{\frac{1+3\nu}{1-\nu}} - \frac{1-2\nu}{1-\nu} \right) c_1 + \left[\frac{\lambda^3 + \frac{1-3\nu}{1-\nu} \lambda^{\frac{1-5\nu}{1-\nu}}}{\sqrt{\lambda^4 + 2\lambda^{\frac{2-6\nu}{1-\nu}}}} - \left(\frac{1-2\nu}{1-\nu} \right) \frac{2}{\sqrt{3}} \right] c_2 + 2 \left\{ \frac{\left(2\lambda^2 + \lambda^{\frac{4\nu}{1-\nu}} \right)^{3c_4}}{\lambda^{\frac{4-8\nu}{1-\nu} c_4}} \left[(3c_4 + 1) \left(\lambda - \frac{\nu}{1-\nu} \lambda^{\frac{1+3\nu}{1-\nu}} \right) - c_4 \frac{1-2\nu}{1-\nu} \left(2\lambda + \lambda^{\frac{1+3\nu}{1-\nu}} \right) \right] - \frac{1-2\nu}{1-\nu} 27^{c_4} \right\} c_3. \tag{28}$$

For the incompressible condition, the Equation (28) with the Poisson's ratio $\nu = 0.5$ is simplified as

$$\tilde{P}_{et} = 2(\lambda - \lambda^{-5})c_1 + \frac{\lambda^3 - \lambda^{-3}}{\sqrt{\lambda^4 + 2\lambda^{-2}}}c_2 + 2(3c_4 + 1)(2\lambda^2 + \lambda^{-4})^{3c_4}(\lambda - \lambda^{-5})c_3. \quad (29)$$

Efforts have been made to reduce shear stretches, increase uniform normal stretches, and minimize branching stability issues by using the bubble inflation technique, square, circular, and cruciform specimens.

3. Applications of Isotropic CSE Model

The isotropic CSE model will be applied to curve-fit experimental data. The experimental data used to identify constitutive parameters will be selected based on the quality of tests at different deformation modes. The quality of the experimental characterizations in the three deformation modes can be evaluated and compared by the three key factors: Accuracy, stretch magnitude, and deformation stability. Uniaxial tension test gives the best accuracy, greatest stretch magnitude, and the best deformation stability among the three most commonly used tests. The equibiaxial tension test produces the worst accuracy, smallest stretch magnitude, and poorest deformation stability due to greater value of $\sqrt{I_2}$ or shear deformation. The quality of a pure shear test falls in between those of uniaxial tension and equibiaxial tension tests. Thus, the isotropic CSE model in uniaxial tension Mode (21) will be selected to curve fit the corresponding experimental data, identifying constitutive parameters. Furthermore, the predictability of the isotropic CSE model will be examined through unfitted experimental data in pure shear and equibiaxial tension modes.

For the isotropic CSE model, five constitutive parameters in general need to be identified based on a uniaxial tension test along with kinematic characterization data by a nonlinear optimization algorithm. The Poisson's ratio, the first constitutive parameter, can be independently determined by fitting the uniaxial kinematic relation test with the Poisson function in uniaxial tension mode. Among the remaining four constitutive parameters of the isotropic CSE Model (21), three parameters c_1 , c_2 , and c_3 are linear and only c_4 is nonlinear, making the identification of constitutive parameters unique. A trial-and-error on digit (TED) method is used to determine c_4 and a linear least square (LLSQ) method is used to resolve the remaining three constitutive parameters c_1 , c_2 , and c_3 simultaneously. Thus, the TED-LLSQ method will be used to identify the constitutive parameters for the isotropic CSE model since it is numerically accurate, stable, and efficient. Nevertheless, multiple solutions are often obtained for stored energy functionals with at least two nonlinear constitutive parameters. With the multiple combinations of constitutive parameters, the predictions of constitutive and finite element models could be quantitatively hugely different as mentioned by Destrade *et al.* (2022) [9].

3.1. Identification of Constitutive Parameters

For $c_4 = A.BCDEFGH \in (0,10)$ example, the TED method starts with ones dig-

it, *i.e.*, $A = \{1, 2, 3, 4, 5, 6, 7, 8, 9\}$. In the nine trials, the number with the smallest error between model and experiment is obtained as A. The number of trials on each of the following digits after the decimal point is 18 instead of 9 since the current number, A.B, could be either larger or smaller than the previous number, A, and a zero digit has been previously tested. Then, there are eighteen trials for $B = \{\pm 0.1, \pm 0.2, \pm 0.3, \pm 0.4, \pm 0.5, \pm 0.6, \pm 0.7, \pm 0.8, \pm 0.9\}$ while adding A.B. together. Then, for $C = 10^{-1}B$ eighteen trials is conducted while adding up to A.BC. A similar process will continue to be executed for sequentially smaller digits up to the desired digit. In this example, the last desired digit is the ten-millionths $H = 10^{-6}B$ while the final decimal number A.BCDEFGH is constructed and solved with only 153 trials.

After the c_4 is obtained at a certain iteration by the TED method, the LLSQ method will be used to resolve the remaining three constitutive parameters c_1 , c_2 , and c_3 simultaneously. The total error between the model and experiment is given by

$$\epsilon = \sum_{j=1}^n \left[P_{ut}(\lambda_j) - P_{ut}^{exp}(\lambda_j) \right]^2, \tag{30}$$

where $P_{ut}^{exp}(\lambda_j)$ represents experimental nominal stress data as a function of stretch λ_j in uniaxial tension tests and n is the total number of pairs of experimental data. The error, ϵ , can be minimized with the following conditions

$$\frac{\partial \epsilon}{\partial c_k} = 0, \quad k = 1, 2, 3. \tag{31}$$

Substituting (21) into (30), and Applying (31) yields the following quasi-linear system

$$\sum_{j=1}^n \begin{bmatrix} A_j A_j & B_j A_j & C_j A_j \\ A_j B_j & B_j B_j & C_j B_j \\ A_j C_j & B_j C_j & C_j C_j \end{bmatrix} \begin{bmatrix} c_1 \\ c_2 \\ c_3 \end{bmatrix} = \sum_{j=1}^n \begin{bmatrix} P_{ut}^{exp}(\lambda_j) A_j \\ P_{ut}^{exp}(\lambda_j) B_j \\ P_{ut}^{exp}(\lambda_j) C_j \end{bmatrix} \tag{32}$$

where the vectors A_j , B_j , and C_j with the known parameters c_4 and ν for the isotropic CSE model in uniaxial tension mode are defined as

$$A_j = 2 \left[\lambda_j - 2\nu \lambda_j^{-2\nu-1} - (1-2\nu) \right], \tag{33}$$

$$B_j = 2 \left[\frac{(1-\nu) \lambda_j^{1-2\nu} - \nu \lambda_j^{-4\nu-1}}{\sqrt{2\lambda_j^{2-2\nu} + \lambda_j^{-4\nu}}} - \frac{1-2\nu}{\sqrt{3}} \right], \tag{34}$$

and

$$C_j = 2 \left\{ \left(\lambda_j^2 + 2\lambda_j^{-2\nu} \right)^{3c_4} / \lambda_j^{(2-4\nu)c_4} \left[(3c_4 + 1) \left(\lambda_j - 2\nu \lambda_j^{-2\nu-1} \right) - c_4 (1-2\nu) \left(\lambda_j + 2\lambda_j^{-2\nu-1} \right) \right] - (1-2\nu) 27^{c_4} \right\}. \tag{35}$$

3.2. Incompressible Constitutive Modeling

The uniaxial tension, pure shear, and equibiaxial tension tests of incompressible

S4035A TPE have been conducted. The uniaxial tension test data of S4035A TPE is used to fit the isotropic CSE Model (22). Numerically Solving (32), along with the Vectors (33), (34), (35), and $\nu = 0.5$, simultaneously obtains the four constitutive parameters as $c_1 = 0.0970449$ MPa , $c_2 = 0.0848708$ MPa , $c_3 = 5.4486398 \times 10^{-7}$ MPa and $c_4 = 0.9251924$. The comparison between the isotropic CSE model and uniaxial tension test of S4035A TPE is shown in **Figure 1(a)**. To further examine the predictability of the isotropic CSE model, the four constitutive parameters identified from fitting the uniaxial tension test data have been submitted into both the pure shear Model (25) and the equibiaxial tension Model (29). The predictions of pure shear and equibiaxial tension tests are shown in **Figure 1(b)** and **Figure 1(c)**, respectively.

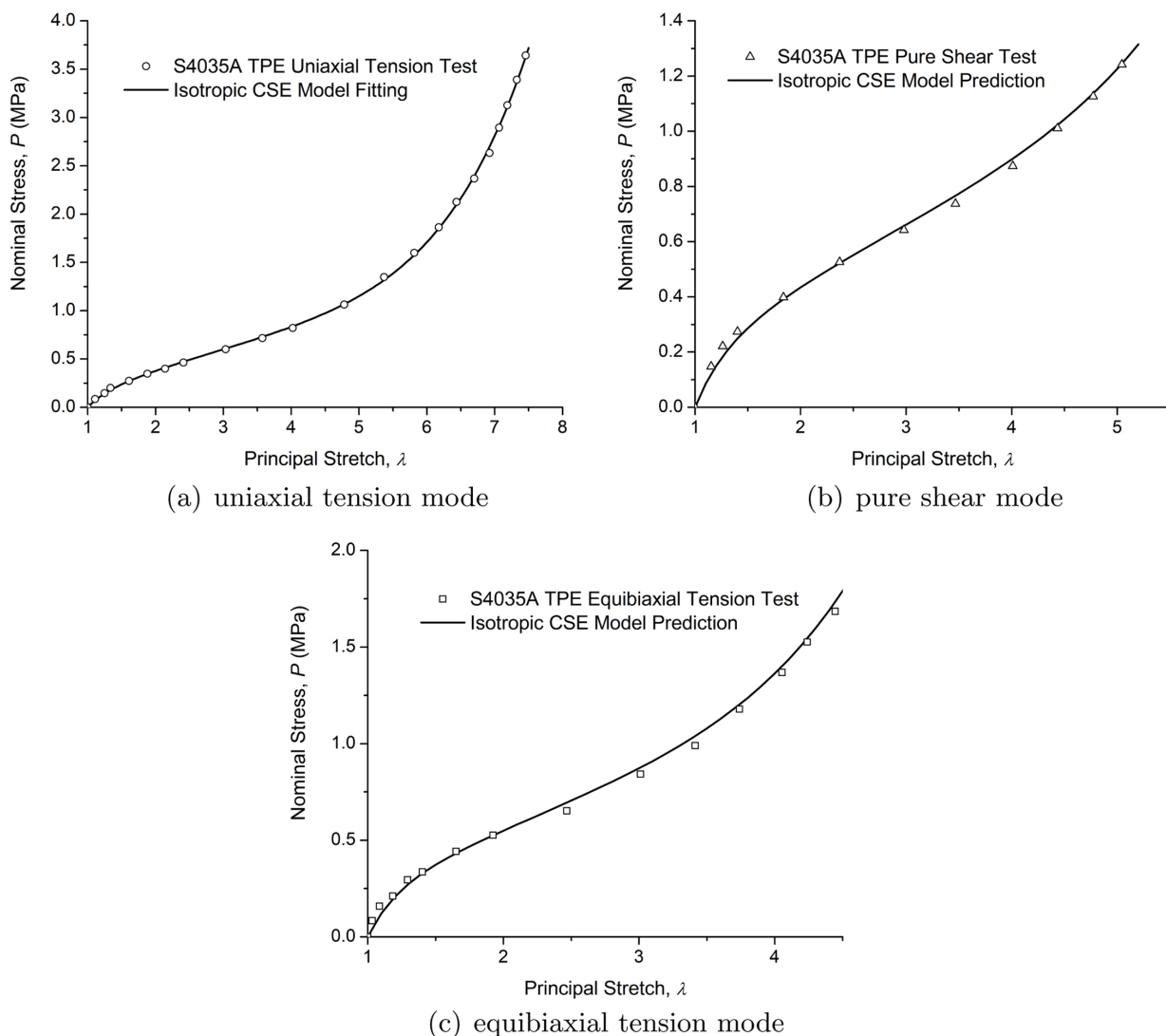


Figure 1. Comparison between experimental test data and isotropic CSE model.

3.3. Compressible Constitutive Modeling

The uniaxial tension, pure shear, and equibiaxial tension tests of compressible

synthetic rubber, along with the detailed kinematic characterizations, have been conducted by Storåkers (1986) [10]. With the kinematic characterizations: lateral contractions as a function of uniaxial stretch, $\lambda_2(\lambda_1)$ and $\lambda_3(\lambda_1)$ data, are used to fit the UT Poisson function. As a result, the Poisson's ratio, $\nu = 0.49122$, is obtained and the fittings of kinematic relationships are shown in **Figure 2(b)**. The uniaxial tension test data of the synthetic rubber is then used to fit the isotropic CSE Model (21). Numerically Solving (32), along with the Vectors (33), (34), (35), and $\nu = 0.49122$, simultaneously obtains the four constitutive parameters as $c_1 = 0.0066309$ MPa, $c_2 = 0.0687864$ MPa, $c_3 = 5.2466927 \times 10^{-5}$ MPa and $c_4 = 0.9733049$. The comparison between the isotropic CSE model and the uniaxial tension test of the synthetic rubber is shown in **Figure 2(a)**. To further examine the predictability of the isotropic CSE model, the five constitutive parameters identified from fitting the uniaxial tension test data have been submitted into both the pure shear Model (24) and the equibiaxial tension Model (28). The predictions of pure shear and equibiaxial tension tests are shown in **Figure 2(c)** and **Figure 2(e)**, respectively. Furthermore, The predictions of kinematic charcaterizations by the PS and ET Poisson functions in pure shear and equibiaxial tension modes are shown in **Figure 2(d)** and **Figure 2(f)**, respectively.

4. Discussion

For the isotropic CSE constitutive model, the five constitutive parameters uniquely identified with uniaxial tension test data for incompressible S4035A TPE and compressible synthetic rubber are listed in **Table 1**.

Table 1. Constitutive parameters of isotropic CSE model for uniaxial tension test.

Material	ν	c_1 (MPa)	c_2 (MPa)	c_3 (MPa)	c_4
S4035A TPE	0.50000	0.0970449	0.0848708	5.4486398×10^{-7}	0.9251924
Synthetic Rubber	0.49122	0.0066309	0.0687864	5.2466927×10^{-5}	0.9733049

4.1. On Incompressible Results

The experimental characterizations of vulcanized natural rubber with 8 phr sulfur have been conducted by Treloar (1944) [11]. Treloar's experiments with all specimens taken from a single sheet of the material are usually used as a benchmark for evaluating constitutive models of incompressible isotropic hyperelastic materials. The isotropic CSE constitutive modeling of Treloar's test data has already been conducted by Zhao (2021) [12].

The uniaxial tension, pure shear, and equibiaxial tension tests of S4035A TPE have been modeled by the isotropic CSE functional with the constant constitutive parameter $c_4 = 1$ [5]. The current models with variable constitutive parameter c_4 achieve better fitting on uniaxial tension test data, better prediction on

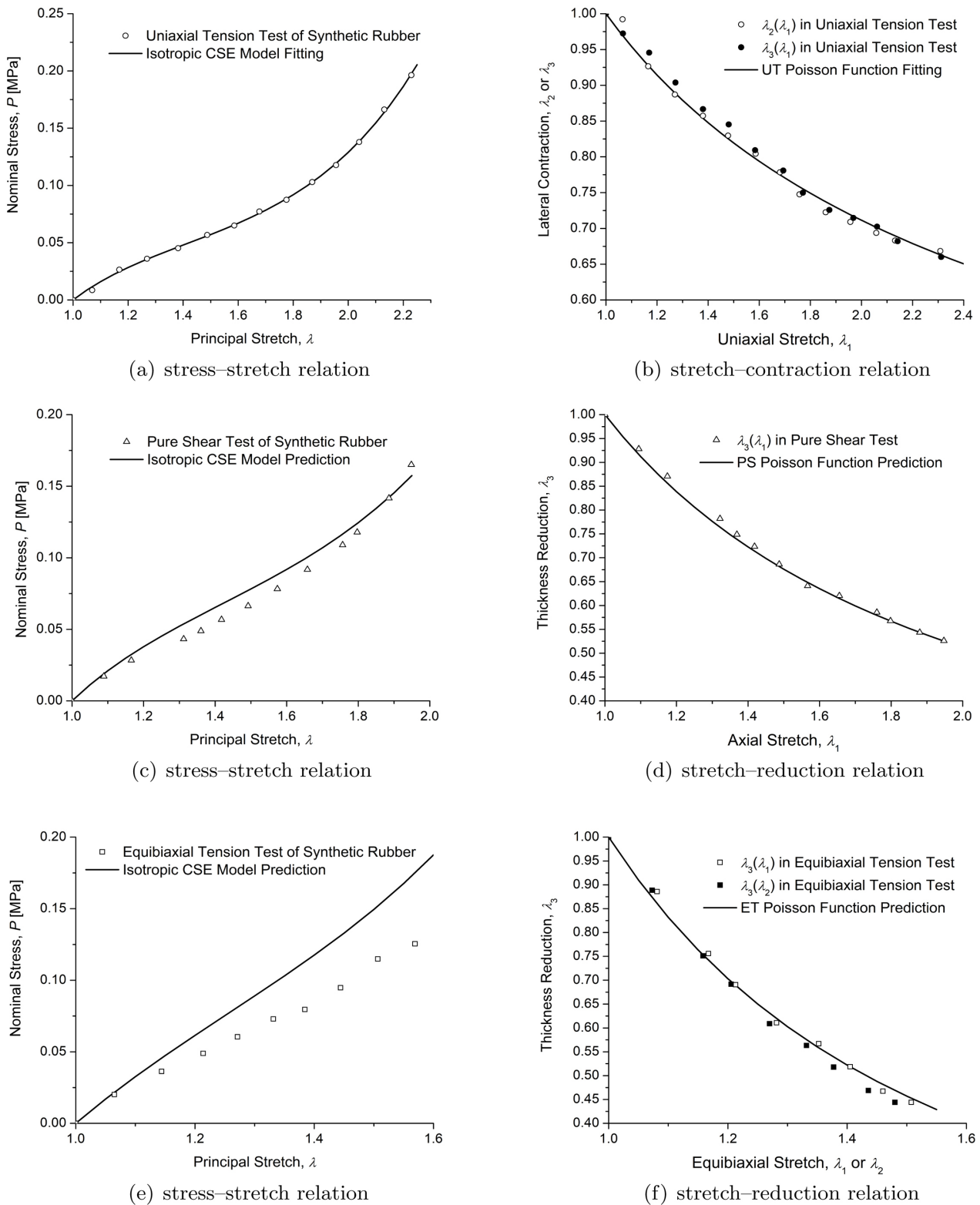


Figure 2. Comparison between experimental test data and isotropic CSE model.

pure shear test data, and particularly better prediction on equibiaxial tension test data of S4035A TPE.

4.2. On Compressible Results

The uniaxial tension, pure shear, and equibiaxial tension tests of compressible natural rubber, along with the detailed kinematic characterizations, conducted by Storåkers (1986) [10] have been studied with the isotropic CSE functional partially combined with the Poisson functions [6].

The Poisson functions can be applied to accurately model and predict available kinematic characterization data for compressible rubberlike materials in different deformation modes [4] [10] [13] [14]. The isotropic CSE functional is fully combined with the Poisson functions to formulate general constitutive equations in the three different deformation modes for compressible as well as incompressible rubberlike materials. The uniaxial tension, pure shear, and equibiaxial tension tests of compressible synthetic rubber, along with the detailed kinematic characterizations, conducted by Storåkers (1986) [10] have been studied with the current isotropic CSE constitutive model. The kinematic characterizations of lateral contractions as a function of uniaxial stretch, $\lambda_2(\lambda_1)$ and $\lambda_3(\lambda_1)$ data, fit the UT Poisson function accurately, indicating a good isotropic behavior of the synthetic rubber shown in **Figure 2(b)**. The kinematic characterizations in pure shear and equibiaxial tension modes are accurately predicted by the PS Poisson function and the ET Poisson function shown in **Figure 2(d)**, and **Figure 2(f)**, respectively. Furthermore, the isotropic CSE constitutive model accurately fits the uniaxial tension test data and predicts the pure shear and equibiaxial tension test data shown in **Figure 2(a)**, **Figure 2(c)**, and **Figure 2(e)**, respectively. The equibiaxial tension test data, however, is underestimated probably due to branching stability issues.

Uniaxial tension mode along with other deformation modes needed to experimentally characterize compressible rubberlike materials may be optimally selected. With advancements in technologies, experimental characterizations for compressible rubberlike materials with better accuracy, greater deformation range, and better stability are still needed. The benchmark tests for compressible rubberlike materials as good as Treloar's tests for incompressible rubberlike materials are yet to appear.

4.3. Principles for Constitutive Equations

The principles of determinism, objectivity, physical consistency, material symmetry, and local actions with dimensional consistency, existence or well-posedness, and equipresence are required for constitutive equations as summarized by Oden (2011) [15].

The principle of physical consistency is crucial since constitutive equations cannot violate the physical principles of mechanics. Furthermore, physical models possess predictive capability while empirical models are approximate mathematical fits to a set of experimental measurements and have no power of prediction as emphasized by Ashby (1992) [16]. Therefore, a constitutive model should be formulated based on the physical principles of mechanics such as the

conservation of energy, the conservation of mass, and the Clausius-Duhem inequality, making it a physical model.

The partial differential equation of isotropic CSE functional was formulated based on the conservation of energy, generally solved by the Lie group method, and specifically solved by the differential geometry. For modeling physically possible nonlinear elastic deformations, the normal stretch, shear stretch, and ellipsoidal stretch deformations are selected based on the curvatures of deformations. Therefore, a constitutive model should be formulated with geometrically meaningful deformations.

The three terms of the isotropic CSE functional are all in the same order of magnitude, λ^2 , since the three terms are asymptotically equal, representing normal stretch, shear stretch, and ellipsoidal stretch deformations. This unique feature of the isotropic CSE functional makes it an isomorphism functional. The thermal deformation does not affect the form of the isotropic CSE constitutive model for structural deformation, which was one kind of material isomorphism assumed by Noll (1972) [17]. Therefore, the finite thermoelastic CSE functional preserves the structure of symmetry in Ψ (16) as studied by Zhao (2023) [18]. In the principle of objectivity, the form of a constitutive model must be invariant under changes in the frame of reference: they must be independent of the observer. As an augmentation, the form of a constitutive model must be invariant under changes in environmental temperature: they must be independent of the environment.

The ideal model is a fully physical model. Constitutive modelings of rubber-like materials are complicated: A fully physical treatment, however, may not be possible. In uniaxial tension deformation as an example, a uniaxial load causes a uniaxial stretch, resulting in lateral contractions. The kinematic relations of two orthogonal lateral contractions with uniaxial stretch cannot be theoretically established based on the conservation of energy since the axial load does not do any work on lateral contraction deformations. For nonlinear finite elastic deformations, the Poisson functions are applied to fit and predict the kinematic relationships at different deformation modes as suggested by Blatz and Ko (1962) [4]. The Poisson's ratio can be independently determined by fitting the UT kinematic relationship tests by the UT Poisson function. Therefore, the isotropic CSE functional, fully combined with the empirical Poisson functions, is applied to constitutively model and predict nonlinear finite elastic deformations for rubberlike materials.

Last but not least, the existence or well-posedness of practical problems in the principle of local action for a constitutive model is required. A constitutive model should have solutions to properly-posed boundary and initial-value problems. Similarly, the constitutive parameters of a constitutive model should be uniquely identified. Otherwise, multiple solutions are often resolved for the constitutive model with two or more nonlinear constitutive parameters. With the multiple combinations of constitutive parameters, the predictions of constitutive models

could be quantitatively widely different as pointed out by Destrade *et al.* (2022) [9].

5. Conclusions

The isotropic CSE functional is fully combined with the Poisson functions to model and predict nonlinear finite elastic deformations in different deformation modes for compressible as well as incompressible rubberlike materials. Experimental characterization requirements in most commonly used uniaxial tension, pure shear, and equibiaxial tension tests are briefly described. The constitutive parameters are uniquely identified by the TED-LLSQ method for the isotropic CSE constitutive model. The principles for constitutive equations are summarized and augmented.

The isotropic CSE constitutive model accurately fits the uniaxial tension test data and accurately predicts the pure shear and equibiaxial tension test data for incompressible S4035A TPE. Furthermore, the isotropic CSE constitutive model accurately fits the uniaxial tension test data and predicts the pure shear and equibiaxial tension test data and the Poisson functions accurately fit and predict the kinematic relation test data for the compressible synthetic rubber.

The isotropic CSE functional, fully combined with the Poisson functions or other kinematic relationships if necessary, will be extensively applied to model and predict mechanical behaviors of compressible as well as incompressible rubberlike materials.

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Conflicts of Interest

The author declares no conflicts of interest to the publication of this paper.

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