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# A new $I_1$-based hyperelastic model for rubber elastic materials

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ABSTRACT

In this Note, we propose a new hyperelastic model for rubber elastic solids applicable over the entire range of deformations. The underlying stored-energy function is a linear combination of the $I_1$-based strain invariants $\varphi(I_1;\alpha) = (I_1^\alpha - 3^\alpha)/(\alpha 3^{\alpha-1})$, where $\alpha$ is a real number. The predictive capabilities of the model are illustrated via comparisons with experimental data available from the literature for a variety of rubbery solids. In addition, the key theoretical and practical strengths of the proposed stored-energy function are discussed.



## 1. Introduction

The Neo-Hookean stored-energy function

$$W = \begin{cases} \frac{\mu}{2}(I_1 - 3) = \frac{\mu}{2}(\lambda_1^2 + \lambda_2^2 + \lambda_3^2 - 3) & \text{if } \lambda_1\lambda_2\lambda_3 = 1 \\ +\infty & \text{otherwise} \end{cases} \tag{1}$$

where $\mu$ stands for the small-strain shear modulus, $I_1$ is the first principal invariant of the right Cauchy–Green deformation tensor $\mathbf{C}$, and $\lambda_1$, $\lambda_2$, and $\lambda_3$ are the associated principal stretches of $\mathbf{C}^{1/2}$, is known to describe reasonably well the mechanical response of rubbery materials at small and moderate deformations. In addition, its very simple mathematical structure has permitted the construction of closed-form solutions for many fundamental boundary-value problems [1], including problems of homogenization [2,3]. For these reasons, expression (1) is generally regarded as the simplest valid prototype for rubber elastic materials.

Since its derivation in the 1940's [4], numerous refinements of relation (1) have been proposed in the literature which give better agreement with experimental data and, in some cases, do capture the limiting chain extensibility of rubber elastic materials at large stretches; the interested reader is referred to the paper of Vahapoglu and Karadeniz [5] for a fairly complete catalogue of hyperelastic constitutive models of rubber proposed between 1930 and 2003. Many of these "refined" models, however, are of considerable mathematical complexity, contain a number of material parameters of uncertain physical value, and/or provide but a somewhat marginally better fit to the experimental data than the simple Neo-Hookean relation (1). In this regard, the objective of this Note is to construct a new (incompressible and isotropic) hyperelastic model that: (i) is simple and amenable enough to analytical closed-form solutions for fundamental boundary-value and homogenization problems, as well as to numerical implementation in commercial finite element packages (e.g., ABAQUS), (ii) contains material parameters which may be given a physical interpretation, and, more importantly, (iii) is able to accurately characterize and predict the mechanical behavior of rubber elastic solids over the entire range of deformations.

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## 2. Proposed constitutive model

With the aim of constructing a *closed-form* stored-energy function that is ultimately of *simple* mathematical structure, we begin by restricting attention to the subclass of incompressible isotropic stored-energy functions that—much like the Neo-Hookean model—depend explicitly on the first principal invariant of the right Cauchy–Green deformation tensor $I_1 = \mathrm{tr}\,\mathbf{C}$ but not on the second invariant $I_2 = 1/2[(\mathrm{tr}\,\mathbf{C})^2 - \mathrm{tr}\,\mathbf{C}^2]$; in the literature, this type of stored-energy functions is referred to as generalized Neo-Hookean. Within this subclass, motivated by the particular $I_1$-based form of relation (1), it is then natural[1] to consider

$$\varphi(I_1;\alpha) = \frac{3^{1-\alpha}}{\alpha}\left(I_1^{\alpha} - 3^{\alpha}\right) \tag{2}$$

where $\alpha$ is a *real number*, as a more appropriate basic measure of strain in place of the less general Neo-Hookean measure $\varphi_{NH}(I_1) = \varphi(I_1;1) = I_1 - 3$. In line with the above reasoning, we then propose a stored-energy function for rubber elastic materials which is a linear combination of the strain measures $\varphi(I_1;\alpha)$, as defined by expression (2), and write[2]

$$W(I_1) = \sum_{r=1}^{M} \frac{3^{1-\alpha_r}}{2\,\alpha_r}\mu_r\left(I_1^{\alpha_r} - 3^{\alpha_r}\right) \tag{3}$$

Here, the integer $M$ denotes the number of terms included in the summation, while $\mu_r$ and $\alpha_r$ $(r = 1, 2, \ldots, M)$ are *real-valued* material parameters that need to be determined ultimately from macroscopic experiments (or possibly from microstructural considerations).

### 2.1. Mathematical simplicity

Similar to any other *closed-form* stored-energy function of the first invariant $I_1$ (e.g., the Gent model [6]), the proposed expression (3) embodies the basic elegance and simplicity of incompressible isotropic hyperelasticity. This is particularly evident in the calculation of the associated Cauchy stress tensor, $\mathbf{T}$, and incremental tangent modulus, $\boldsymbol{\mathcal{L}}$, quantities needed in the analytical study of boundary-value problems, as well as in the numerical implementation in finite element programs. Specifically, the Cauchy stress tensor resulting from (3) is simply

$$\mathbf{T} = \frac{\partial W}{\partial \mathbf{F}}\mathbf{F}^T - p\mathbf{I} = \left(\sum_{r=1}^{M} 3^{1-\alpha_r}\mu_r I_1^{\alpha_r-1}\right)\mathbf{F}\mathbf{F}^T - p\mathbf{I} \tag{4}$$

where $\mathbf{F}$ stands for the deformation gradient tensor, $p$ denotes the arbitrary hydrostatic pressure associated with the incompressibility constraint, and $\mathbf{I}$ is the identity operator in the space of second-order tensors (i.e., $I_{ij} = \delta_{ij}$, with $\delta_{ij}$ denoting the Kronecker delta). The principal Cauchy stresses are given by

$$t_i = \left(\sum_{r=1}^{M} 3^{1-\alpha_r}\mu_r I_1^{\alpha_r-1}\right)\lambda_i^2 - p \quad (i = 1, 2, 3) \tag{5}$$

Moreover, the incremental tangent modulus takes the following simple form

$$\boldsymbol{\mathcal{L}} = \frac{\partial^2 W}{\partial \mathbf{F}^2} = 2\left(\sum_{r=1}^{M} 3^{1-\alpha_r}(\alpha_r - 1)\mu_r I_1^{\alpha_r-2}\right)\mathbf{F}\otimes\mathbf{F} + \left(\sum_{r=1}^{M} 3^{1-\alpha_r}\mu_r I_1^{\alpha_r-1}\right)\boldsymbol{\mathcal{I}} \tag{6}$$

where the symbol $\otimes$ has been introduced to denote the tensor product (i.e., $(\mathbf{F}\otimes\mathbf{F})_{ijkl} = F_{ij}F_{kl}$) and $\boldsymbol{\mathcal{I}}$ stands for the identity operator in the space of fourth-order tensors (i.e., $\mathcal{I}_{ijkl} = \delta_{ik}\delta_{jl}$). In passing, besides its simplicity, it is worth remarking that the explicit expression (6) is valid for *all* deformations as it stands. This is in contrast to the corresponding expressions associated with stored-energy functions that are written in terms of the principal stretches $\lambda_1$, $\lambda_2$, and $\lambda_3$ (e.g., the Ogden model [7]). Indeed, stretch-based tangent incremental moduli exhibit singularities whenever $\lambda_i = \lambda_j$ for $i \neq j$ and therefore, appropriate limiting expressions must be used in place of the singular general expressions in these cases.

[1] Another natural alternative is to consider $\psi(I_1;\alpha) = (I_1 - 3)^{\alpha}$, but this measure—as opposed to (2)—has the disadvantage that it does not linearize properly in the limit of small deformations.

[2] Here and subsequently, the unbounded branch of the stored-energy function for non-isochoric deformations will be omitted for notational simplicity.

### 2.2. Physical significance of material parameters

In spite of the fact that the proposed stored-energy function (3) has been constructed in a heuristic manner, the underlying material parameters, $\mu_r$ and $\alpha_r$, may be given a physical interpretation. To see this, we make contact with the non-Gaussian statistical mechanics model recently put forward by Beatty [8], for which the macroscopic Cauchy stress tensor has the form

$$\mathbf{T} = \Psi(I_1)\mathbf{F}\mathbf{F}^T - p\mathbf{I} \tag{7}$$

where $\Psi(I_1)$ is a certain microstructural function (see relation (7.2), together with (7.1) in [8]) that contains information about the statistical distribution of the underlying polymeric chains in the material.

Note that the proposed constitutive relation (4) for the Cauchy stress is precisely of the general form (7), indicating that the constitutive constants $\mu_r$ and $\alpha_r$ can potentially have physical significance (see [9] for a similar assertion regarding the Gent model). It is beyond the aim of this Note to carry out an in-depth study of the connections between (4) and the general form of the Beatty model (7), and so we limit ourselves to compare (4) with a special case of (7) which is particularly well suited to reveal the fact that the material parameters $\mu_r$ and $\alpha_r$ in (3) can indeed be given a physical interpretation. Such a special case of the Beatty model (4) is essentially nothing more than the Arruda–Boyce 8-chain model [10] (but see Section 6 in [8] for the relevant derivation and for some caveats):

$$\mathbf{T}_{AB} = \frac{nkT}{3}\frac{\mathcal{L}^{-1}(\sqrt{I_1/3\eta})}{\sqrt{I_1/3\eta}}\mathbf{F}\mathbf{F}^T - p\mathbf{I} \tag{8}$$

In this last expression, the variables $n$, $k$, $T$, $\eta$ denote, respectively, the chain density, the Boltzmann constant, the absolute temperature, and the number of chain links in a representative single chain, while $\mathcal{L}^{-1}$ denotes the inverse Langevin function. The stored-energy function associated with (8) *cannot* be written in closed form, but a convenient power series representation can be written as follows

$$W_{AB}(I_1) = \frac{nkT}{2}\sum_{r=1}^{\infty} C_r\left(I_1^r - 3^r\right) \tag{9}$$

where the parameters $C_r$ are known functions of the number of chain links $\eta$ (e.g., $C_1 = 1$, $C_2 = 1/(10\eta)$, $C_3 = 11/(525\eta^2), \ldots$).

A glance at (9) suffices to recognize that by choosing $\mu_r = 3^{r-1}rnkTC_r$, $\alpha_r = r$, and $M = \infty$, the proposed stored-energy function (3) reduces identically to the Arruda–Boyce 8-chain model. Thus, for this particular choice, the material constants $\mu_r$ and $\alpha_r$ do have physical value, as they can be directly associated with a Langevin-type statistical distribution of the underlying polymeric chains. From a broader perspective, the special power series structure of expression (9) does also suggest that the parameters $\mu_r$ and $\alpha_r$ in the similar but much richer functional structure (3) can be associated with more general (than Langevin) forms of statistical distributions of the polymeric chains. The verity of such a physical interpretation of the material constants $\mu_r$ and $\alpha_r$ is worth studying in future work.

### 2.3. Predictive capabilities over the entire range of deformations

In the sequel, we demonstrate the ability of the proposed model to characterize and predict the mechanical response of rubber elastic materials for large ranges of deformations. From a *qualitative* point of view, it is first appropriate to remark that expression (3) admits the following polynomial representation

$$W(I_1) = \sum_{i=1}^{\infty}\frac{3^{1-i}}{2i!}\left[\sum_{r=1}^{M}\left(\prod_{j=1}^{i-1}(\alpha_r - j)\right)\mu_r\right](I_1 - 3)^i \tag{10}$$

from which it is a simple matter to deduce that in the limit as $I_1 \to 3$ expression (3) reduces to

$$W(I_1) = \sum_{r=1}^{M}\frac{\mu_r}{2}(I_1 - 3) + \sum_{r=1}^{M}\frac{\alpha_r - 1}{12}\mu_r(I_1 - 3)^2 + O\left((I_1 - 3)^3\right) \tag{11}$$

Thus, in the *small-deformation regime*, the model response is seen to be independent of the material parameters $\alpha_r$ and, what is more, identically Neo-Hookean with

$$\sum_{r=1}^{M}\mu_r = \mu \tag{12}$$

In the *moderate-deformation regime*, by contrast, the model response departs from Neo-Hookean behavior, as it accounts for higher-order polynomial terms in the invariant $I_1 - 3$. Incidentally, this feature—namely, accounting for higher-order terms

in $I_1 - 3$—has been shown (see, e.g., [11]) to work well in describing the behavior of rubbery materials at moderate deformations. Turning attention now to the *large-deformation regime* (i.e., $I_1 \gg 3$), it is plain to recognize that expression (3) is strongly dependent on the parameters $\alpha_r$ in this range of deformations. An immediate consequence of this strong dependence is that the proposed model has the ability to capture the typical limiting chain extensibility of rubbers at large stretches. This is easy to realize, for instance, by setting a sufficiently large value for some $\alpha_r$, which would drastically penalize increases in deformation in the large-deformation regime. In summary, the proposed constitutive model appears to have the right qualitative features to be able to characterize the behavior of rubber elastic materials over the entire range of deformations.

The *quantitative* capabilities of the stored-energy function (3) to model rubber elastic materials are illustrated next via comparisons with experimental data available from the literature for 3 different types of rubbery solids: (i) a vulcanized rubber (Treloar, 1944 [12]), (ii) a silicone rubber (Meunier, 2008 [13]), and (iii) a commercial elastomer from the tire company Michelin (Lahellec et al., 2004 [14]). For demonstration purposes, it suffices to consider two terms ($M = 2$) in expression (3)—the one-term ($M = 1$) stored-energy function can be shown to lead to good predictions for small and moderate deformations but it cannot simultaneously capture the limiting chain extensibility at large stretches—and therefore we write

$$W(I_1) = \frac{3^{1-\alpha_1}}{2\alpha_1}\mu_1\big(I_1^{\alpha_1} - 3^{\alpha_1}\big) + \frac{3^{1-\alpha_2}}{2\alpha_2}\mu_2\big(I_1^{\alpha_2} - 3^{\alpha_2}\big) \tag{13}$$

For convenience, we also write down explicitly the stress-strain relations resulting from the two-term stored-energy function (13) for:

- *Uniaxial loading* ($\lambda_1 = \lambda$, $\lambda_2 = \lambda_3 = \lambda^{-1/2}$ with $t_2 = t_3 = 0$):

$$S_{un} = \lambda^{-1}t_1 = \frac{\mathrm{d}W}{\mathrm{d}\lambda} = \frac{\lambda^3 - 1}{2\lambda + \lambda^4}\sum_{r=1}^{2} 3^{1-\alpha_r}\mu_r\big(\lambda^2 + 2\lambda^{-1}\big)^{\alpha_r} \tag{14}$$

- *Biaxial loading* ($\lambda_1 = \lambda_2 = \lambda$, $\lambda_3 = \lambda^{-2}$ with $t_2 = t_1$, $t_3 = 0$):

$$S_{bi} = \lambda^{-1}t_1 = \frac{1}{2}\frac{\mathrm{d}W}{\mathrm{d}\lambda} = \frac{\lambda^6 - 1}{\lambda + 2\lambda^7}\sum_{r=1}^{2} 3^{1-\alpha_r}\mu_r\big(2\lambda^2 + \lambda^{-4}\big)^{\alpha_r} \tag{15}$$

- *Pure shear* ($\lambda_1 = \lambda$, $\lambda_2 = \lambda^{-1}$, $\lambda_3 = 1$ with $t_2 = 0$):

$$S_{ps} = \lambda^{-1}t_1 = \frac{\mathrm{d}W}{\mathrm{d}\lambda} = \frac{\lambda^4 - 1}{\lambda + \lambda^3 + \lambda^5}\sum_{r=1}^{2} 3^{1-\alpha_r}\mu_r\big(\lambda^2 + \lambda^{-2} + 1\big)^{\alpha_r} \tag{16}$$

- *Simple shear* ($\lambda_1 = (\gamma + \sqrt{\gamma^2 + 4})/2$, $\lambda_2 = (\gamma - \sqrt{\gamma^2 + 4})/2$, $\lambda_3 = 1$):

$$S_{ss} = \frac{\mathrm{d}W}{\mathrm{d}\gamma} = \frac{\gamma}{\gamma^2 + 3}\sum_{r=1}^{2} 3^{1-\alpha_r}\mu_r\big(\gamma^2 + 3\big)^{\alpha_r} \tag{17}$$

In the above expressions (14)–(17), we have introduced the scalar Piola–Kirchhoff stress measures $S_{un}$, $S_{bi}$, $S_{ps}$, and $S_{ss}$ for consistency with the experimental stress measurements, which are given in terms of force per unit undeformed area of cross section.

We begin by considering the classical experimental data of Treloar [12] for the uniaxial, biaxial, and pure shear loading of a vulcanized rubber. Our immediate task is to find values for the four material parameters $\alpha_1$, $\alpha_2$, $\mu_1$, and $\mu_2$ in (13) that give the best fit to the data of any one of Treloar's experiments. Because of the larger range of deformations considered—with a maximum reported stretch of $\lambda_{\max} \approx 7.5$ (see Fig. 1(a))—we choose to fit Treloar's uniaxial experiment. After enforcing proper linearization (so that, in this case, $\mu = \mu_1 + \mu_2 = 0.27$ MPa), the best-fitting values for $\alpha_1$, $\alpha_2$, $\mu_1$, and $\mu_2$ obtained by means of a least-squares fit in this case are displayed in Table 1. The resulting stress-stretch model response (14), together with the experimental data, is presented in Fig. 1(a). Note that the correlation between the model and the experiment is excellent for the entire range of deformations considered.

Using the material parameters in Table 1—which, again, have been generated from the uniaxial tension data only—we can readily make use of the constitutive relations (15) and (16) to *predict* the response for biaxial tension and pure shear loadings. The results are presented in Fig. 1(b). The model prediction for pure shear is seen to be in good agreement with the experimental measurements. The agreement between the model prediction for biaxial tension and the experimental data is fair.

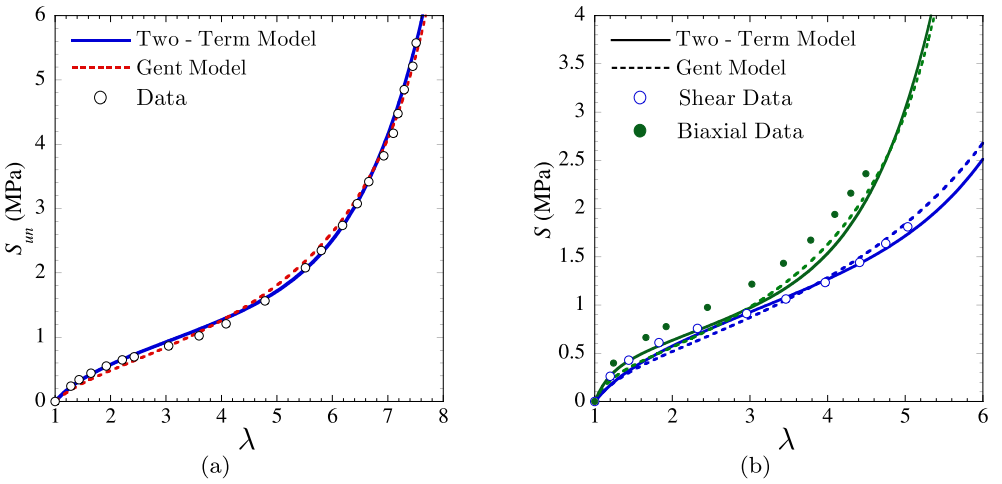


**Fig. 1.** Two-term model (13) and the Gent model (18), with the material parameters of Table 1, compared with the data of Treloar (1944) [12] for: (a) uniaxial tension, and (b) biaxial tension and pure shear of a vulcanized rubber.

**Table 1**
Material parameters fitted to the uniaxial data of Treloar (1944) [12].

| | | |
|---|---|---|
| Two-term model (13) | $\alpha_1 = 1.08$ | $\mu_1 = 0.2699$ MPa |
| | $\alpha_2 = 4.40$ | $\mu_2 = 0.00001771$ MPa |
| Gent model (18) | $J_m = 85.91$ | $\mu = 0.27$ MPa |

We now turn attention to the more comprehensive set of experiments of Meunier et al. [13] for the uniaxial compression/tension, biaxial tension, pure shear compression, and pure shear tension loadings of a silicone rubber. Similar to the preceding case, our first task is to find values for the four material parameters $\alpha_1$, $\alpha_2$, $\mu_1$, and $\mu_2$ in (13) that give the best fit to the data of any one of the experiments while being consistent with a proper linearization in the limit of small deformations. Once more, because of the larger range of deformations considered, we choose to fit the uniaxial data (including both compression and tension). The best-fitting values for $\alpha_1$, $\alpha_2$, $\mu_1$, and $\mu_2$ obtained in this case are given in Table 2. The resulting stress-stretch model response (14) is presented in Fig. 2(a), where it is seen to be in excellent agreement with the experiment for the entire range of deformations considered.

Making use of the material parameters in Table 2, we can readily employ the constitutive relations (15) and (16) to *predict* the response for biaxial tension, pure shear compression, and pure shear tension. The results are presented, together with the corresponding experimental measurements, in Fig. 2(b), (c), and (d), respectively. Here again, the model predictions are seen to exhibit a remarkably good agreement with the experimental data for all three loading conditions.

Finally, we consider the experiments of Lahellec et al. [14] for the uniaxial tension and simple shear of a commercial elastomer synthesized by the tire company Michelin. Following the approach of computing the values for the material parameters in (13) from the single experiment with largest applied deformation, in this case, we generate the values of $\alpha_1$, $\alpha_2$, $\mu_1$, and $\mu_2$ given in Table 3 by fitting the uniaxial tension measurements of Lahellec et al. [14]. In spite of exhibiting a distinctive nonlinearity at relatively small deformations, the uniaxial response of this commercial elastomer is shown in Fig. 3(a) to be well characterized by the theoretical stress-stretch relation (14). Using the material parameters in Table 3, it is then a simple matter to employ the constitutive relation (17) to *predict* the simple shear response of the elastomer. The result, which is plotted in Fig. 3(b), is seen once more to be in good agreement with the experimental measurements.

In summary, the above examples have demonstrated the ability of the proposed stored-energy function (13) to model, fairly accurately, the mechanical response of different classes of rubbery solids under a wide variety of loading conditions. In this regard, it is important to re-emphasize that for all three elastomers examined in Figs. 1–3, the underlying material parameters, $\mu_r$ and $\alpha_r$, were obtained by (least-squares) fitting only the data of one experiment (uniaxial loading), and that these same fitted parameters were then used to *predict* the behavior of the elastomers under different loading conditions. This approach—of fitting just one experiment[3]—has served to highlight: (i) the truly predictive capabilities of the model, and (ii) the ease and numerical robustness to determine the values of its material parameters $\mu_r$ and $\alpha_r$.

[3] Of course, the material parameters in (13) can alternatively be computed by fitting all the available experiments (not just uniaxial loading), which may possibly lead to an improved overall fit to the experimental data.

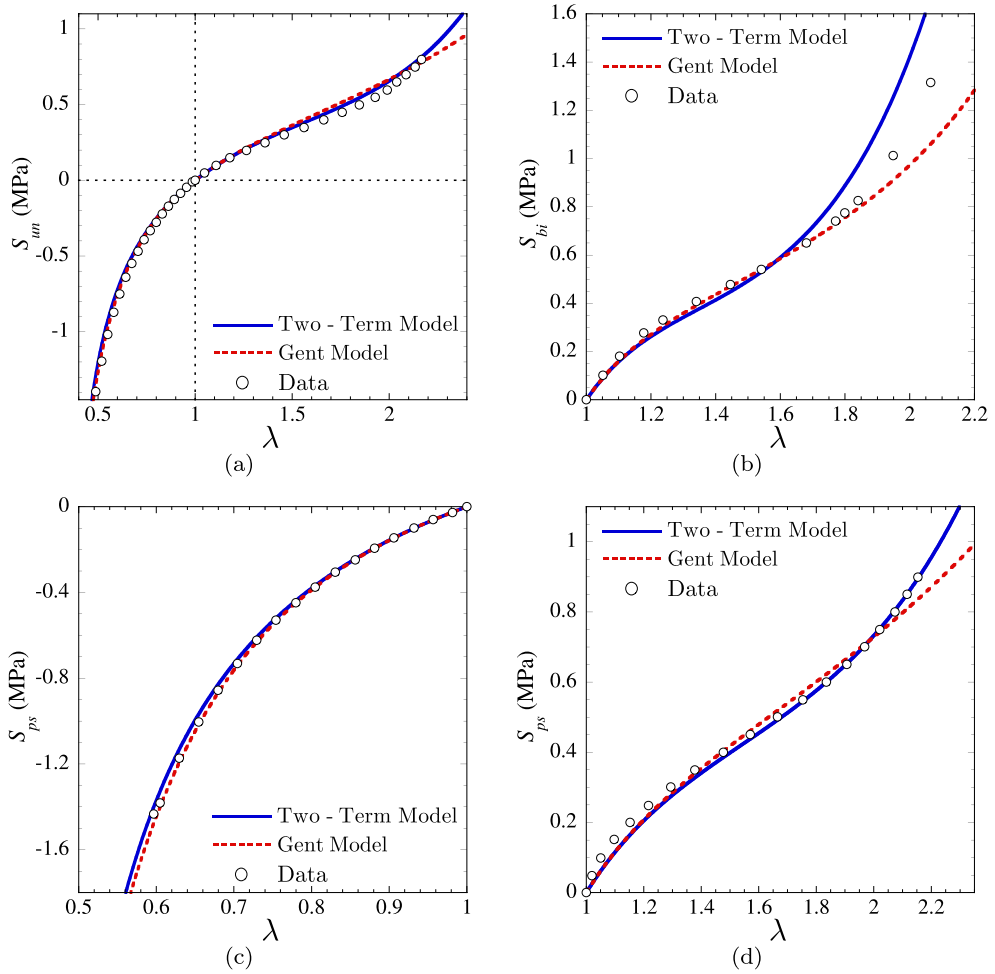


**Fig. 2.** Two-term model (13) and the Gent model (18), with the material parameters of Table 2, compared with the data of Meunier et al. (2008) [13] for: (a) uniaxial compression and tension, (b) biaxial tension, (c) pure shear compression, and (d) pure shear tension of a silicone rubber.

**Table 2**
Material parameters fitted to the uniaxial data of Meunier et al. (2008) [13].

| | | |
|---|---|---|
| Two-term model (13) | $\alpha_1 = 3.837$ | $\mu_1 = 0.032$ MPa |
| | $\alpha_2 = 0.559$ | $\mu_2 = 0.3$ MPa |
| Gent model (18) | $J_m = 15.42$ | $\mu = 0.332$ MPa |

### *2.4. Comparison with existing $I_1$-based models*

In the literature, there are a number of $I_1$-based constitutive relations for rubber elastic materials [5]. Among these, arguably the most widely utilized are the Gent model:

$$W_G(I_1) = -\frac{\mu J_m}{2} \ln\left[1 - \frac{I_1 - 3}{J_m}\right] \tag{18}$$

where $\mu$ and $J_m$ are material constants, and the already introduced Arruda–Boyce model (9). Despite their seemingly different functional forms, the Gent and Arruda–Boyce models can be shown to be essentially identical to each other [15], with the practical difference that the Gent—as opposed to the Arruda–Boyce—model is *closed-form* and therefore simpler to use and more numerically robust [16].

In order to gain further insight into the new stored-energy function (13), the response of the popular Gent constitutive relation (18)—which, again, is essentially identical to that of (9)—has been included in Figs. 1–3 for comparison purposes. As for the two-term relation (13), the 2 material parameters $\mu$ and $J_m$ in (18) are obtained by fitting the relevant uniaxial data, after enforcing proper linearization. The results are given in Tables 1, 2, and 3, respectively, for the vulcanized rubber

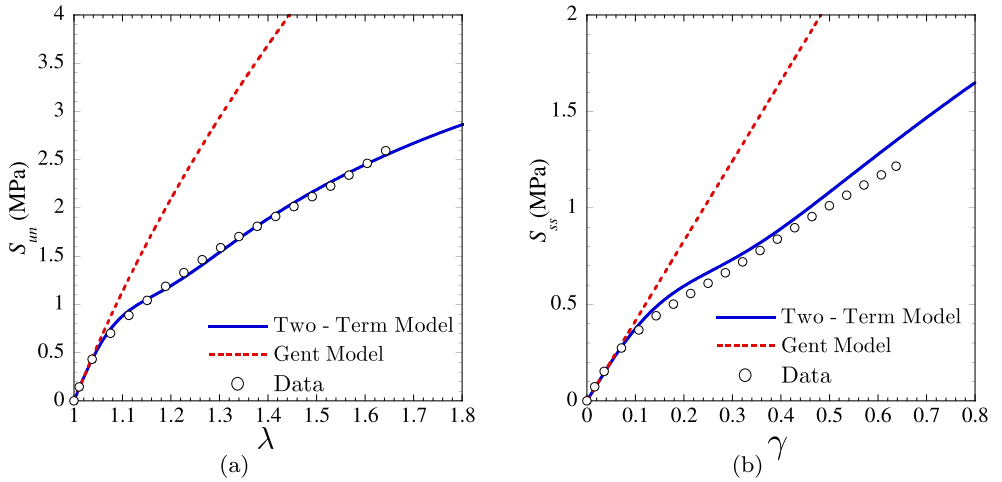


**Fig. 3.** Two-term model (13) and the Gent model (18), with the material parameters of Table 3, compared with the data of Lahellec et al. (2004) [14] for: (a) uniaxial tension and (b) simple shear of a commercial rubber from Michelin.

**Table 3**
Material parameters fitted to the uniaxial data of Lahellec et al. (2004) [14].

| | | |
|---|---|---|
| Two-term model (13) | $\alpha_1 = 0.6$ | $\mu_1 = 2.228$ MPa |
| | $\alpha_2 = -68.73$ | $\mu_2 = 1.919$ MPa |
| Gent model (18) | $J_m = 1.91 \times 10^{15}$ | $\mu = 4.147$ MPa |

of Treloar [12], the silicone rubber of Meunier et al. [13], and the Michelin elastomer of Lahellec et al. [14]. These same fitted parameters are then utilized to predict the response of the rubbers under the various loading conditions.

Fig. 1 shows that the Gent model leads to very similar results to those generated by the two-term relation (13) for the uniaxial, biaxial, and pure shear loading of the vulcanized rubber. For the silicone rubber, it is observed from Fig. 2 that the Gent predictions are in good agreement with the experiments, but the agreement is noticeably not as good as that of the model (13). Finally, Fig. 3 shows that the Gent model—as opposed to the new model (13)—is unable to characterize the behavior of the Michelin elastomer for uniaxial loading, and, in addition, fails to predict its response under simple shear beyond the small-deformation regime.

It is plain from Fig. 3 (and, although to a lesser extent, from Fig. 2 as well) that the main reason behind the overall superior correlation between the two-term model (13) and the experimental data is not merely due to the fact that it contains more material constants than the Gent model—namely, 4 constants ($\alpha_1$, $\alpha_2$, $\mu_1$, and $\mu_2$) versus 2 ($\mu$ and $J_m$)—but rather to its richer functional structure. Indeed, it is particularly evident from Fig. 3 that the functional form of the Gent model is too restrictive to be able to characterize the strongly nonlinear response of some elastomers, even for uniaxial loading. This favorable comparison provides further evidence supporting the use of expression (2) as a fundamental $I_1$-based measure of strain.

## 3. Final remarks

### 3.1. Constitutive restrictions on $\mu_r$ and $\alpha_r$

The mathematical structure of the proposed stored-energy function (3) naturally embraces the notion of polyconvexity introduced by Ball [17] to prove existence theorems in finite elasticity. In this connection, it is not difficult to show that necessary and sufficient conditions for (3) to be strictly polyconvex read simply as

$$W'(I_1) > 0 \quad \text{and} \quad W'(I_1) + 2I_1 W''(I_1) > 0 \tag{19}$$

where

$$W'(I_1) = \sum_{r=1}^{M} \frac{3^{1-\alpha_r}}{2} \mu_r I_1^{\alpha_r - 1}, \qquad W''(I_1) = \sum_{r=1}^{M} \frac{3^{1-\alpha_r}(\alpha_r - 1)}{2} \mu_r I_1^{\alpha_r - 2} \tag{20}$$

Unfortunately, necessary and sufficient conditions on the parameters $\mu_r$ and $\alpha_r$ that ensure that (3) is strictly polyconvex do not exist. But it is straightforward to deduce from (19) that the conditions

$$\mu_r > 0 \quad \text{and} \quad \alpha_r > \frac{1}{2} \quad (r = 1, 2, \ldots, M) \tag{21}$$

are sufficient.

Given that (strict) polyconvexity implies (strict) rank-one convexity [17], the restrictions (19) are also sufficient to ensure the strong ellipticity of the stored-energy function (3). To see this connection more explicitly, we spell out the necessary and sufficient conditions for the strong ellipticity of (3) (see, e.g., Section 4 in [18]):

$$W'(I_1) > 0 \quad \text{and} \quad W'(I_1) + 2\big(I_1 - \lambda_i^2 - 2\lambda_i^{-1}\big)W''(I_1) > 0 \quad (i = 1, 2, 3) \tag{22}$$

where it is recalled that the principal stretches $\lambda_i > 0$ $(i = 1, 2, 3)$ satisfy the incompressibility constraint $\lambda_1\lambda_2\lambda_3 = 1$. After recognizing the string of inequalities $I_1 > I_1 - \lambda_i^2 - 2\lambda_i^{-1} \geqslant 0$ $(i = 1, 2, 3)$, it is evident that the conditions of strict polyconvexity (19)—and therefore the simpler parametric restrictions (21)—indeed imply the conditions of strong ellipticity (22).

Although desirable on a mathematical basis, the constitutive restriction of polyconvexity (19) has not yet been given a strict physical interpretation and therefore its enforcement is arguable. On the other hand, the strong ellipticity condition (22) must be enforced in general, since—consistent with experimental evidence on neat rubbery solids—it entails physically that localized deformations (e.g., shear bands) cannot develop in the solid.

Making contact with the two-term relation (13) utilized in Section 2.3, it is relevant to remark that the inequalities (21) are satisfied by the material parameters $(\alpha_1, \alpha_2, \mu_1, \mu_2)$ generated from the experimental data of Treloar [12] and Meunier et al. [13], as given in Tables 1 and 2, respectively, and therefore the resulting stored-energy functions satisfy conditions (22). On the other hand, the material parameters obtained from the data of Lahellec et al. [14] (see Table 3) do not satisfy the sufficient inequalities (21), but it is straightforward to verify that they lead to a stored-energy function (13) that is fully consistent with the strong ellipticity conditions (22), as expected.

### 3.2. Generalizations

The proposed stored-energy function (3) constitutes a practical platform from which to account for more levels of complexity to model rubbery solids. Below, we discuss some of these generalizations.

From the pioneering work of Rivlin and Saunders [19] (see also Chapters 10 and 11 in the monograph by Treloar [20] and references therein), it is well known that the response of rubbery solids depends not only on the first principal invariant $I_1$, but also on the second invariant $I_2 = 1/2[(\mathrm{tr}\,\mathbf{C})^2 - \mathrm{tr}\,\mathbf{C}^2]$. The dependence on $I_2$ is, however, much weaker and that is the main motivation to neglect $I_2$ effects altogether as a first approximation. Nevertheless, at the expense of sacrificing mathematical simplicity, it may be of interest to explore generalizations of the stored-energy function (3) that incorporate dependence on $I_2$. A plausible generalization that has proved fruitful in the related context of the Gent model could consist in adding the logarithmic term $\ln(I_2/3)$ to $W$ [21]. Alternatively, given the theoretical and practical virtues of the strain measure (2), it may also be of interest to explore

$$W(I_1, I_2) = \sum_{r=1}^{M} \frac{3^{1-\alpha_r}}{2\alpha_r}\mu_r\big(I_1^{\alpha_r} - 3^{\alpha_r}\big) + \sum_{s=1}^{N} \frac{3^{1-\beta_s}}{2\beta_s}\nu_s\big(I_2^{\beta_s} - 3^{\beta_s}\big) \tag{23}$$

where $\beta_s$ and $\nu_s$ $(s = 1, 2, \ldots, N)$—much like $\alpha_r$ and $\mu_r$ $(r = 1, 2, \ldots, M)$—are *real-valued* material parameters.

Starting with the classical work of Bridgman [22], a variety of experiments have established that rubbery solids are not exactly incompressible (and, in some cases, they may actually be quite compressible). In this regard, there are a number of ways in which compressibility effects can be readily incorporated into the stored-energy function (3). For instance, a simple compressible version of (3) is given by

$$W(I_1, J) = \sum_{r=1}^{M} \frac{3^{1-\alpha_r}}{2\alpha_r}\mu_r\big(I_1^{\alpha_r} - 3^{\alpha_r}\big) - \sum_{r=1}^{M} \mu_r \ln J + \frac{\mu'}{2}(J-1)^2 \tag{24}$$

Here, $J = \lambda_1\lambda_2\lambda_3$ and the parameter $\mu'$, which agrees with the Lamé constant in the ground state (recall that $\mu = \sum_{r=1}^{M}\mu_r$ corresponds to the other Lamé constant), serves to measure the compressibility of the material. Specifically, note that in the limit of incompressibility as $\mu' \to \infty$, expression (24) reduces identically to the stored-energy function (3), together with the incompressibility constraint $J = \lambda_1\lambda_2\lambda_3 = 1$. In passing, it is fitting to record that a form similar to (24) has recently been utilized to successfully model the compressible polybutadiene phase in a certain class of SBS triblock copolymers [23].

Examples of other phenomena that may be added to the stored-energy function (3) include Mullins, hysteresis, as well as rate and thermal effects. The works of Miehe [24] and Ogden and Roxburgh [25] may be relevant here.

### 3.3. Application to reinforced and porous elastomers

Often times, rubbery solids are reinforced with fibers and/or particles to improve their stiffness. For use in packaging, cushioning, and energy absorption applications, some elastomers are also weakened with voids. In a recent effort, Lopez-Pamies and Ponte Castañeda [26] have put forward a novel homogenization theory that aims to model such classes of soft

heterogeneous materials by incorporating direct dependence on the constitutive behavior of the underlying constituents (e.g., the matrix and fibers in a fiber-reinforced material) and the microstructure (e.g., the size, shape, and distribution of the fibers). As a first application of the theory, constitutive models have been recently derived for various classes of fiber-reinforced [27–29] and porous [30–32] materials with constituents that are characterized by any $I_1$-based stored-energy function of choice. With the help of (3), these homogenization results can now be utilized to model large classes of rubber elastic composites.

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