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A three-parameter structurally motivated robust constitutive model for isotropic incompressible unfilled and filled rubber-like materials

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ABSTRACT

A structurally motivated three-parameter strain energy function W is presented in this paper for application to the constitutive modelling of various types of incompressible isotropic rubber-like materials, from unfilled to filled rubbers, polymers and soft tissues. A particular objective of the current work is to achieve another step towards devising a *comprehensive* model for application to an assortment of incompressible isotropic hyperelastic materials, not limited only to a specific subset type of rubbers or polymers. To this end, the application of the model to extant datasets from various types of rubber-like materials is presented, via simultaneous fitting of the model to the considered deformation modes. The modelling results are then compared with those of a selected set of established three-parameter models in the literature, including a generalised neo-Hookean strain energy function and another two models which include an I_2 term. It will be shown that the proposed model most favourably captures the datasets for all unfilled, filled rubber and polymer specimens considered. Finally, the model is applied to the deformation datasets of a soft tissue, namely the human brain, for which the (one-term) Ogden model is currently known to provide the best fit. It will be demonstrated that the application of the Ogden model will lead to loss of convexity, while our proposed model provides a better fit, with lower relative errors, and remains convex over the deformation domain. By way of the presented examples and applications, it is concluded that the proposed model provides a reliable degree of *robustness* for a suitable application to constitutive modelling of *various types* of rubber-like materials.

1. Introduction

In assessing the proficiency of the existing hyperelastic constitutive models, ranging from the seminal review of Marckmann and Verron (2006) to more recent studies by Destrade et al. (2017) and Dal et al. (2021), three major criteria may be identified for choosing a suitable model for application to the mechanics of isotropic rubber-like materials. These three criteria are: (i) ability of the model to provide favourable fits to various deformation datasets of the same rubber specimen via a single set of model parameter values obtained by simultaneous fitting of the model to the datasets (Marckmann and Verron, 2006; Dal et al., 2021); (ii) possessing a minimal number of model parameters (Destrade et al., 2017; Dal et al., 2021); and (iii) ideally having molecular structural roots (Marckmann and Verron, 2006; Dal et al., 2021) so that the model parameters would enjoy a degree of physical *objectivity* and mathematical underpinning.

Each of the foregoing identified traits have important implications beyond just the matter of providing a good fit to a dataset. The ability of a model to capture various deformation modes of a specimen via a single set of model parameter values speaks to the *robustness* of that model, and whether the obtained model parameters can be entrusted to reliably represent the true constitutive characteristics of the subject specimen. As identified by Marckmann and Verron (2006), even some of the most celebrated hyperelastic models such as the Ogden (1972) model do not always admit a single set of parameter values to capture the various deformations of the same material. An added layer to the notion of *robustness* of a model manifests itself in the capability of the said model in capturing the mechanical behaviour of *various* rubber-like materials, as opposed to be only applicable to a specific subset type. An eminent example in this regard, as will be discussed further in this work, is finding a suitable model that can be applied to both unfilled and filled rubbers, and/or polymers that exhibit discerning mechanical features.

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As suggested by a number of studies (e.g., Lopez-Pamies, 2010; Nunes, 2011; Nunes and Moreira, 2013; Moreira and Nunes, 2013; Araújo and Nunes, 2020), finding such a general model is rare in the current literature.

Possessing a minimal number of model parameters is another important feature which has direct mathematical and mechanical *stability* implications. Ogden et al. (2004) provide a detailed analysis on how an excessive number of model parameters may impede obtaining a unique optimal fit and parameter values. Yeoh (1997) also observed that the additional degrees of freedom introduced by considering unwarranted extra parameters over what is constitutively admissible for a specimen will only lead to a model that fits better the experimental errors. Accordingly, Yeoh (1997) stipulates that strain energy functions with higher number of model parameters are more prone to being *unstable*; i.e., lead to unrealistic behaviours outside the range of the experimental data. This undesirable feature also underlines the importance of the third identified trait, namely the advantage in having a model with molecular/structural basis which in turn ensures a degree of physical *objectivity* and meaning for the embodied model parameters (e.g., Anssari-Benam and Bucchi, 2021).

To this end, studies seeking to 'rank' the capability of the existing hyperelastic models (e.g., Marckmann and Verron, 2006; Destrade et al., 2017; Dal et al., 2021) have mostly employed the canonical datasets of Treloar (1944) and Kawabata et al. (1981), which of course pertain to (vulcanised) natural rubber specimens. Identifying a model which satisfies the three foregoing criteria becomes even more challenging if modelling the mechanical behaviour of filled rubbers is also added to the requirements. Filled rubbers, as well as some polymeric materials, exhibit an apparent deformation-softening response, reflected by a downward concavity in stress versus deformation curves (see, e.g., the datasets on (carbon) filled rubber by Lahellec et al. (2004) and on some polymeric and silicone samples by Nunes, 2011; Nunes and Moreira, 2013; Moreira and Nunes, 2013 and Araújo and Nunes, 2020). An overview of the shear-softening behaviour demonstrated by these datasets in the context of simple shear has been presented in a recent study by Anssari-Benam and Horgan (2021), where we highlighted that such a behaviour cannot be expected to be captured by many of the existing models in the literature. Indeed, the only model known to the authors that has been shown capable of capturing this behaviour is that due to Lopez-Pamies (2010), using up to two terms with four parameters in relation to the dataset of Lahellec et al. (2004). However, if applications require more terms of this model to be included, then this model enters the territory identified by Ogden et al. (2004) in respect of finding an optimal fit and the uniqueness of the obtained model parameter values. It therefore appears that a structurally motivated model that contains fewer model parameters will be more advantageous in this regard.

A preliminary effort at presenting a strain energy function that is applicable to both unfilled and filled rubbers, is structurally motivated, contains two constitutive parameters, and provides favourable fits to many deformation datasets of the specimens via simultaneous fitting was commenced in Anssari-Benam and Horgan (2021), with a particular focus on simple shear deformation. We will further develop that study here to include uniaxial, biaxial and pure shear deformations. Our overall goal is to develop a new class of robust constitutive models that are suitable for application to *various types* of rubber-like materials and many deformation modes of the specimens, reflected by the quality of fits with the experimental data and improved relative errors. Accordingly, here we propose a new hyperelastic strain energy function with only *three* model parameters which provides the *generality* for application to both natural and filled rubbers, with simultaneous fitting to various deformation datasets of the specimens. Favourable applications to other rubber-like materials such as soft tissues and (silicone) polymers will also be demonstrated, by way of highlighting that the application capability of the proposed model is *not limited* to rubbers. The considered model, as will be shown in the sequel, is a generalisation of the parent model utilised by Anssari-Benam and Bucchi (2018, 2021), which was recently developed by Anssari-Benam (2021) as a [1/1] Padé approximant of the non-Gaussian inverse Langevin function $\mathscr{L}^{-1}$.

Accordingly, in Section 2 we present and summarise the theoretical preliminaries for the proposed model. The application of the model to datasets of unfilled and filled rubbers, as well as polymeric silicones will be demonstrated in Section 3. A brief comparison with other three-parameter models in the literature including those which contain an I_2 (the second invariant of the left Cauchy-Green deformation tensor $\mathbf{B}$) term will also be presented. These models include the cubic model due to Yeoh (1993), modification proposed by Pucci and Saccomandi (2002) to the original Gent (1996) model, also referred to as the Gent-Gent model, and the model proposed by Carroll (2011). One motivation for including the latter two models that contain an I_2 term is to show that inclusion of such a dependence does not *always* lead to improved fitting with experimental data. Clear advantages of the proposed model over those models will be demonstrated and discussed. In addition, we also consider a specific soft-tissue application via a dataset pertaining to the deformation of the human brain (cortex) tissue due to Budday et al. (2020b). The impetus for considering this particular dataset is to show the improved fits achieved by the proposed model compared with the popular model currently considered in the literature, namely the one-term Ogden model. It will be shown that the application of the new model to this dataset does not lead to a loss of convexity as is the case for the one-term Ogden model. Concluding remarks are made in Section 4.

2. The three-parameter model

The mathematical form of the strain energy function W of interest here was derived and presented by Anssari-Benam (2021):

$$W = \frac{3(n-1)}{2n}\mu N\left[\frac{1}{3N(n-1)}(I_1-3) - \ln\left(\frac{I_1-3N}{3-3N}\right)\right], \tag{1}$$

where n, μ and N are the three model parameters to be described in the following, and I_1 is the first invariant of the left Cauchy-Green deformation tensor $\mathbf{B}$. The response function of this model, i.e., $\partial W/\partial I_1$, is a [1/1] Padé approximant in I_1 and is directly related to the rounded Padé approximant of the inverse Langevin function $\mathscr{L}^{-1}$ (Anssari-Benam, 2021). It is therefore noted that this model is structurally motivated, being based on the non-Gaussian statistics of the molecular chains. The model in Eq. (1) is the parent model of that utilised by Anssari-Benam and Bucchi (2018, 2021), where in the special case when $n=3$ that model is recovered. We note here that the model by Anssari-Benam and Bucchi (2018, 2021) is itself a special case of the nonaffine network model first introduced by Davidson and Goulbourne (2013).

The parameter N in model (1) is the number of links, also known as the Kuhn segments, of the constituent molecular chains (see also the review by Puglisi and Saccomandi (2016) for a microstructural interpretation of the parameters derived from the kinetic molecular theory of rubber elasticity), and the parameter n is related to the degree of strain-hardening at a given N. In the original derivation of the model by Anssari-Benam (2021), the numerical value of n is restricted to natural numbers, $n \in \mathbb{N}$, i.e., $n = 1, 2, 3\ldots$. However, phenomenologically there is no reason that n has to be numerically limited as such, and therefore here we generalise this condition to:

$$n \in \mathbb{R}^+, \tag{2}$$

i.e., n is a positive real-valued parameter. The infinitesimal shear modulus μ_0 for this class of model is then:

$$\mu_0 = 2\left(\frac{\partial W}{\partial I_1}\right)_{I_1=3} \Rightarrow \mu_0 = \frac{1}{n}\mu\frac{1-nN}{1-N}, \tag{3}$$

or, alternatively, the parameter μ in the model has the following relationship with the infinitesimal shear modulus μ_0:

$$\mu = \mu_0 \frac{n(1-N)}{1-nN} \ . \tag{4}$$

From Eqs. (3) and (4) it is readily observed that in the limit as $N \to \infty$, then $\mu = \mu_0$ and the model in Eq. (1) simplifies to the neo-Hookean strain energy function:

$$\lim_{N\to\infty} W = \frac{1}{2}\mu_0 (I_1 - 3) \ . \tag{5}$$

Also note that for the case where $n = 1$, we again have $\mu = \mu_0$ and the neo-Hookean model is once more recovered. On re-writing the model in Eq. (1) using the relationship in Eq. (4) in terms of μ_0 we obtain:

$$W = \frac{3N(n-1)(1-N)}{2(1-nN)}\mu_0 \left[\frac{1}{3N(n-1)}(I_1 - 3) - \ln\left(\frac{I_1 - 3N}{3-3N}\right)\right] , \tag{6}$$

which is subjected to the following rational constraints:

$$\mu_0 > 0 \quad , \quad \frac{I_1 - 3N}{3-3N} > 0 \ . \tag{7}$$

While the parameter N is structurally restricted to $N > 1$, the constraint $(7)_2$ does not require such an imposition and therefore, as discussed at length by Anssari-Benam and Horgan (2021), this restriction may be relaxed so that N is treated as a phenomenological real-valued parameter. Furthermore, from Eq. (6) we obtain:

$$W_1 = \frac{\partial W}{\partial I_1} = \frac{1}{2}\mu_0 \left(\frac{1-N}{1-nN}\right)\left(\frac{I_1 - 3nN}{I_1 - 3N}\right) \ . \tag{8}$$

It is well known that to satisfy the so-called empirical inequalities one requires $W_1(I_1) > 0$ (see, e.g., the discussion in Beatty, 1987). To ensure that W_1 in Eq. (8) satisfies the condition $W_1(I_1) > 0$, it is sufficient to require:

$$\begin{cases} \dfrac{1-N}{1-nN} > 0 \ , \\[2ex] \dfrac{I_1 - 3nN}{I_1 - 3N} > 0 \ , \end{cases} \tag{9}$$

or:

$$\begin{cases} \dfrac{1-N}{1-nN} < 0 \ , \\[2ex] \dfrac{I_1 - 3nN}{I_1 - 3N} < 0 \ . \end{cases} \tag{10}$$

Thus, the conditions in (7) must hold together *with either* of (9) or (10). To analyse these conditions, it is convenient to consider the two cases $N > 1$ and $N < 1$ separately:

(i) $N > 1$: It can be easily verified that in this case the condition in $(7)_2$ holds if and only if:

$$I_1 < 3N \ , \tag{11}$$

and that then, the conditions in (9) are satisfied if and only if:

$$N > \frac{1}{n} \ , \ \ I_1 < 3nN \ . \tag{12}$$

Alternatively the conditions (10) hold if and only if:

$$N < \frac{1}{n} \ , \ \ I_1 > 3nN \ . \tag{13}$$

(ii) $N < 1$: It can be easily verified that in this case the condition in $(7)_2$ holds if and only if:

$$I_1 > 3N \ . \tag{14}$$

Then, the constraint in (9) holds if and only if:

$$N < \frac{1}{n} \ , \ \ I_1 > 3nN \ . \tag{15}$$

Alternatively, the constraint in (10) holds if and only if:

$$N > \frac{1}{n} \ , \ \ I_1 < 3nN \ . \tag{16}$$

It is of interest to note here that the model in Eq. (1) is indeed also the *parent* to the Gent (1996) model. We observe from Eq. (6) that in the limit where $n \to \infty$ we have:

$$\lim_{n\to\infty} W = -\frac{1}{2}(3N-3)\mu_0 \ln\left(\frac{I_1 - 3N}{3-3N}\right) \ . \tag{17}$$

By setting the limiting stretch parameter J_m in the Gent model as: $J_m = 3N - 3$, Eq. (17) may readily be re-written to:

$$\lim_{n\to\infty} W = -\frac{1}{2}J_m\mu_0 \ln\left[\frac{I_1 - (J_m + 3)}{-J_m}\right] = -\frac{1}{2}J_m\mu_0 \ln\left[1 - \frac{I_1 - 3}{J_m}\right], \tag{18}$$

which is, of course, the strain energy function of the Gent model. It is therefore observed that the two-parameter Gent model is a special subset of the proposed model in Eq. (1). Note that the response function of the Gent model is a [0/1] Padé approximant in I_1 (see, e.g., Horgan and Saccomandi, 2002 and Beatty, 2008). The response function of the proposed model, on the other hand, is a higher order [1/1] Padé approximant in I_1, and is directly related to the rounded [3/2] Padé approximation of $\mathscr{L}^{-1}$ in terms of the molecular chain stretch λ_c (Anssari-Benam, 2021). These attributes render the proposed model a more accurate representation. See also Anssari-Benam and Horgan (2022a) for the hierarchy of the order of the Padé approximants of the inverse Langevin function for the response functions of some of the existing generalised neo-Hookean limiting chain extensibility models. Even for the special case $n = 3$, i.e., the model in Anssari-Benam and Bucchi (2018, 2021), where the response function is shown by Horgan (2021) to differ from that of the Gent model by just an additive constant, the improved accuracy of the predictions of the former model versus the Gent model has been demonstrated (e.g., see, Anssari-Benam and Bucchi, 2021; Anssari-Benam et al., 2021b). Recent applications of this special model have also been described in Anssari-Benam and Horgan (2022a, 2022b; 2022c). We emphasise that the proposed model (1) is the more generalised, more accurate and the *parent class* of the two-parameter Gent, Anssari-Benam and Bucchi and similar models.

For application to experimental data, which will be presented in Section 3, we now briefly present the stress-deformation relationships for the model in Eq. (1). The Cauchy stress $\mathbf{T}$ for an isotropic incompressible material is given by:

$$\mathbf{T} = -p\mathbf{I} + 2W_1\mathbf{B} - 2W_2\mathbf{B}^{-1} \ , \tag{19}$$

where $\mathbf{B}$ $(=\mathbf{F}\mathbf{F}^{\mathrm{T}})$ is the left Cauchy-Green deformation tensor and $\mathbf{B}^{-1}$ is its inverse, p is the arbitrary Lagrange multiplier enforcing the condition of incompressibility, $\mathbf{I}$ is the identity tensor, and W_1 and W_2 are the partial derivatives of the strain energy function W with respect to the first and second principal invariants of $\mathbf{B}$, which are defined as:

$$I_1 = \lambda_1^2 + \lambda_2^2 + \lambda_3^2 \quad , \quad I_2 = \lambda_1^{-2} + \lambda_2^{-2} + \lambda_3^{-2} \ , \tag{20}$$

with $I_3 = 1$ due to incompressibility so that $\lambda_1\lambda_2\lambda_3 = 1$. From these, it is easily shown that the $T - \lambda$ relationship for uniaxial deformation using model (1) becomes:

$$T_{uni} = \frac{1}{n}\mu\left(\frac{I_1 - 3nN}{I_1 - 3N}\right)\left(\lambda^2 - \frac{1}{\lambda}\right), \tag{21}$$

while that for equi-biaxial deformation is:

$$T_{eq-bi} = \frac{1}{n}\mu\left(\frac{I_1 - 3nN}{I_1 - 3N}\right)\left(\lambda^2 - \frac{1}{\lambda^4}\right). \tag{22}$$

Similarly, for pure shear we have:

$$T_{ps} = \frac{1}{n}\mu\left(\frac{I_1 - 3nN}{I_1 - 3N}\right)\left(\lambda^2 - \frac{1}{\lambda^2}\right), \tag{23}$$

and for the case of simple shear with the amount of shear γ we find:

$$T_{ss} = \frac{1}{n}\mu\left(\frac{I_1 - 3nN}{I_1 - 3N}\right)\gamma. \tag{24}$$

We will use the relationships in Eqs. (21)–(24) in the next Section for simultaneous fitting of the model to the relevant experimental datasets.

3. Applications

As highlighted in Section 2, the proposed model in Eq. (1) is *structurally* motivated, directly connected with the non-Gaussian statistics of the molecular chain network via a [1/1] Padé approximation of the inverse Langevin function $\mathscr{L}^{-1}$. The model also contains *only three* constitutive parameters, which have structural links and interpretations through the infinitesimal shear modulus μ_0, the number of Kuhn segments N and the degree of strain-hardening n at a given N (Anssari-Benam and Bucchi, 2018, 2021; Anssari-Benam, 2021). Therefore, from a theoretical point of view, the proposed model satisfies two out of the three criteria highlighted in the literature, and recounted in the opening statement in Section 1, regarding the characteristics of a suitable hyperelastic model. The remaining feature is the capability to provide a degree of reliable *robustness*, in that different deformations modes of a specimen must ideally be captured via a single set of model parameters, obtained by simultaneous fitting of the model to the data (Marckmann and Verron, 2006; Dal et al., 2021). To this we also suggest a further desirable criterion, with an additional aspect of *robustness*, which is that a model should be able to adequately capture the mechanical behaviour of *various* rubber-like specimens, i.e., not only limited to a specific subset of materials such as, for example, natural rubbers etc. Many, if not most, of the celebrated existing models in the literature have been shown to fail to provide satisfactory fits to filled rubber specimens, very soft biological tissues, or even various vulcanised natural rubbers. For a demonstration of the shortcomings of various models in this regard we refer the reader to numerous contributions including those by Marckmann and Verron (2006), Destrade et al. (2017) and Lopez-Pamies (2010) in relation to natural unfilled and filled rubbers, and Anssari-Benam and Horgan (2021), Anssari-Benam and Bucchi (2018), Budday et al. (2020a; 2020b) and Mihai et al. (2015) in relation to soft tissues, *inter alia*. The models considered in the aforementioned studies are not limited to a specific type (e.g, generalised neo-Hookean etc) or specific inclusion/exclusion of certain invariants etc. Rather, these studies indicate that the functional forms of many, if not most, of the existing models are *not* conducive to the universality required for a satisfactory application to the deformation of various rubber-like materials. Most of the existing models, therefore, are at best suitable for application to a certain subset of rubber-like materials.

Accordingly, in this section we present the results of applying the proposed model to extant experimental datasets from a range of *different types* of rubber-like materials, including natural (vulcanised) rubber, (carbon) filled rubber, silicone-based polymers and, by way of an example, a biological soft tissue. Capturing the deformation of filled rubbers has particularly proved challenging for most models, as filled rubber specimens tend to exhibit a 'softening' effect where the stress-deformation curve possesses a downward concavity. Many models fail to capture this prominent mechanical feature of filled rubbers. By contrast, as will be shown in this section, the proposed model provides favourable fits to the data.

In all the curve-fitting demonstrations that will be presented in the sequel, the model is fitted simultaneously to the various deformations of the specimens to test and demonstrate the capability of the model to provide universal modelling results. The fitting procedure that we employ is similar to our previous undertakings. To characterise the model parameters, optimisation is sought by fitting the $T - \lambda$ (and/or $T - \gamma$ where γ represents the amount of shear) relationships given in Eqs. (21)–(24) simultaneously to the associated deformation datasets. The best fit is achieved by minimising the residual sum of squares (RSS) function defined as: RSS $= \sum_i \left(T^{model} - T^{experiment}\right)_i^2$, where i is the number of data points. The minimisation is performed via an in-house developed code in MATLAB®, using the genetic algorithm (GA) toolbox. The Coefficient of determination R^2 is used as the measure for the goodness of the fits. The presented relative error (%) in the plots is calculated as: $\left|\frac{T^{model} - T^{experiment}}{T^{experiment}}\right| \times 100$.

3.1. Unfilled (vulcanised) rubber

Following the comprehensive reviews carried by Marckmann and Verron (2006) and Dal et al. (2021), standard datasets for unfilled rubbers considered in the comparison of modelling capabilities in the literature are the canonical datasets due to Treloar (1944) and Kawabata et al. (1981). Accordingly, we consider these datasets here too. The tabulated experimental data pertaining to these datasets have been presented in our other studies, see, e.g., Anssari-Benam and Bucchi (2021).

Both these datasets report experimental data on uniaxial, equi-biaxial and pure shear deformation of a vulcanised natural rubber. Eqs. (21)–(23) were therefore simultaneously fitted to each of the two datasets. The plots in Figs. 1 and 2 illustrate the modelling results for the Treloar (1944) and Kawabata et al. (1981) datasets, respectively. The ensuing model parameter values have been listed in Tables 1 and 2, respectively.

It is important to observe that the values of N in Tables 1 and 2 *are* $N > 1$ *so that, in fitting with all the preceding datasets, the model (1) is being tested within its original molecular based framework*. In passing we note that according to Marckmann and Verron (2006), since these two datasets are from specimens of a similar rubber material, an appropriate model should be able to capture both datasets using a similar range of parameter values. It may be observed from Tables 1 and 2 that, while the values of the model parameters are not exactly the same for the two datasets, the similarity in the values of the model parameters for the two

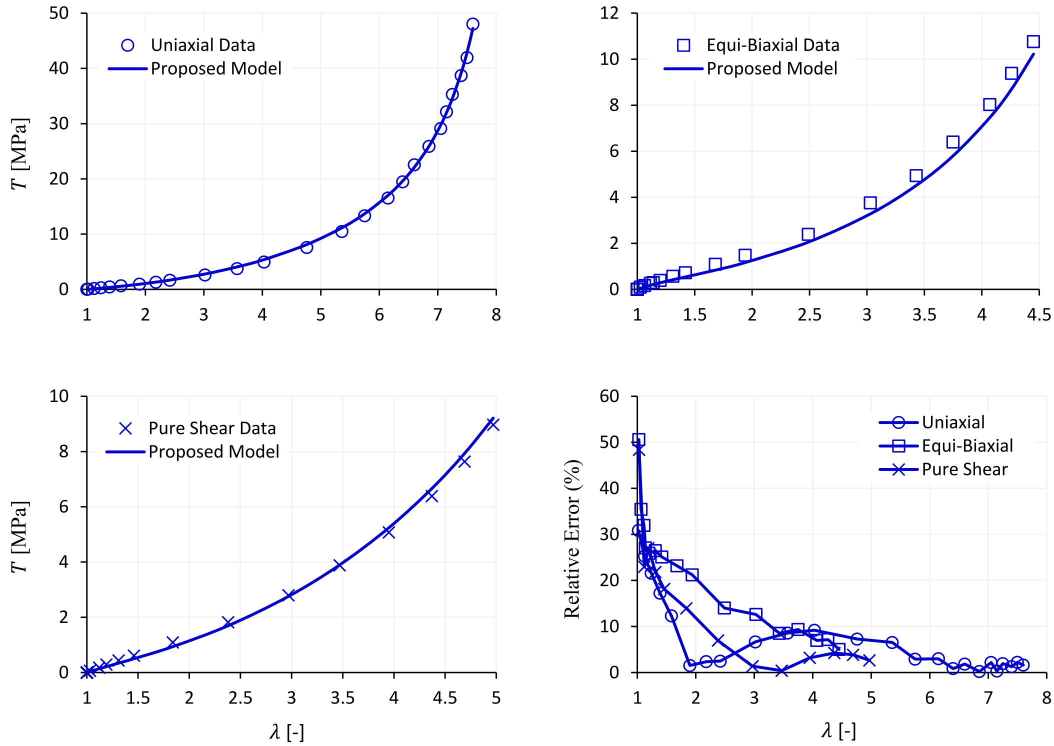

Fig. 1. Modelling results for the Treloar (1944) dataset using the proposed model (1).

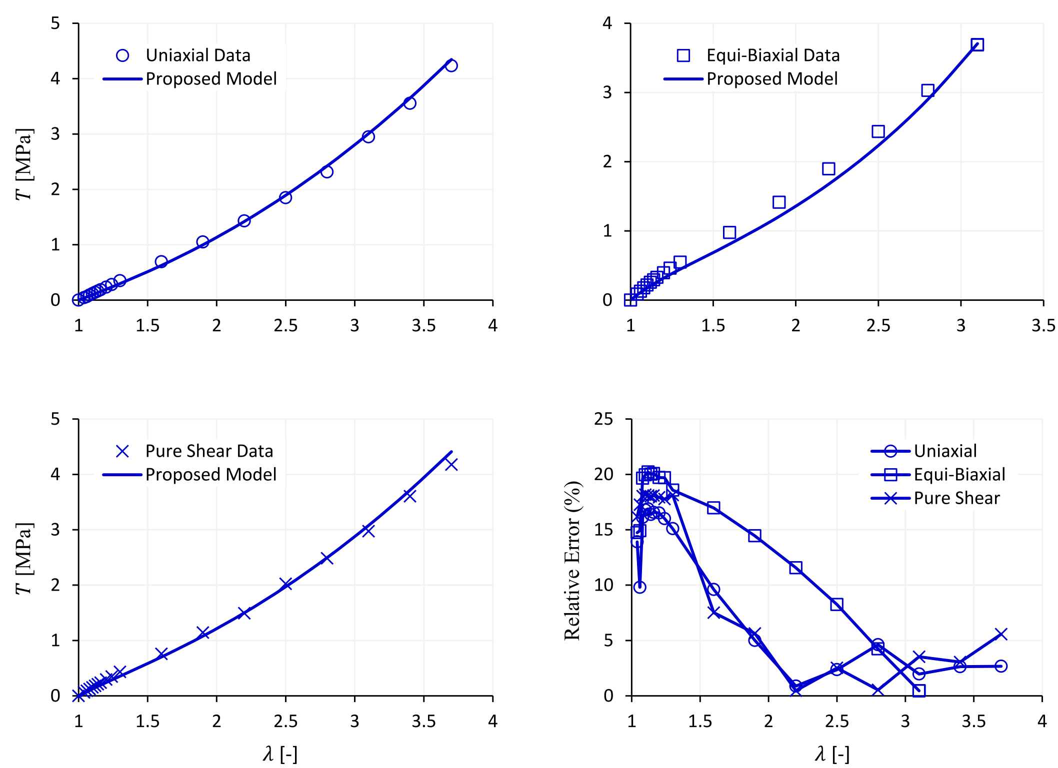

Fig. 2. Modelling results for the Kawabata et al. (1981) dataset using the proposed model (1).

Table 1
Model parameters for the Treloar (1944) dataset using the model (1).

Deformation mode	μ [MPa]	N [-]	n [-]	R^2	μ_0 [MPa][a]
Uniaxial	0.29	24.96	2.09	0.99	0.30
Equi-biaxial				0.99	
Pure shear				0.99	

[a] Calculated on using Eq. (3) with the given values of μ, N and n.

Table 2
Model parameters for the Kawabata et al. (1981) dataset using the model (1).

Deformation mode	μ [MPa]	N [-]	n [-]	R^2	μ_0 [MPa][a]
Uniaxial	0.32	21.17	1.002	0.99	0.32
Equi-biaxial				0.99	
Pure shear				0.99	

[a] Calculated on using Eq. (3) with the given values of μ, N and n.

datasets is noticeable.

At this juncture it may be informative to provide a comparison between the foregoing modelling results and those of some *three-parameter* models already established in the literature. To this effect, we consider the Yeoh (1993) cubic model and the Gent-Gent model (Pucci and Saccomandi, 2002). Both these models contain three parameters, similar to our proposed model in Eq. (1), and thus from a fitting perspective provide a similar degree of freedom. The Gent-Gent model also includes an I_2 term, which as pointed out in the Introduction, will help testing the hypothesis that whether a functional dependence on I_2 *necessarily* leads to an improved fitting with experimental data.

Starting with the cubic model of Yeoh (1993) where the strain energy function is given by:

$$W(I_1) = \mathscr{C}_1(I_1 - 3) + \mathscr{C}_2(I_1 - 3)^2 + \mathscr{C}_3(I_1 - 3)^3, \tag{25}$$

with $\mathscr{C}_1$, $\mathscr{C}_2$ and $\mathscr{C}_3$ being positive model parameters, we note the inferior quality of the fits obtained when the model is simultaneously fitted to the Treloar (1944) dataset. The modelling results are presented in Fig. 3. Comparing the relative errors obtained via the cubic model with that of the proposed model in Fig. 1, it is clear that while both models contain three parameters, the proposed model in Eq. (1) provides a better fit. A similar trend was also observed for Kawabata et al. (1981) data; however, in the interest of brevity we refrain from reproducing the results here.

The Gent-Gent model (Pucci and Saccomandi, 2002) has the following from:

$$W(I_1, I_2) = -\frac{\mu_0}{2} J_m \ln\left(1 - \frac{I_1 - 3}{J_m}\right) + C_2 \ln\left(\frac{I_2}{3}\right), \tag{26}$$

where μ_0, J_m and C_2 are the three model parameters. While this model provides improved fits to the Treloar (1944) data compared with the original Gent model (see, e.g., the results by Pucci and Saccomandi, 2002), it is evident from the plots in Fig. 4 that the Gent-Gent model does not offer an improvement compared with the proposed model in Eq. (1). Indeed, by looking at the relative error plots, we observe that the relative errors arising from the application of the Gent-Gent model are generally higher than those resulting from the proposed model, for equi-biaxial

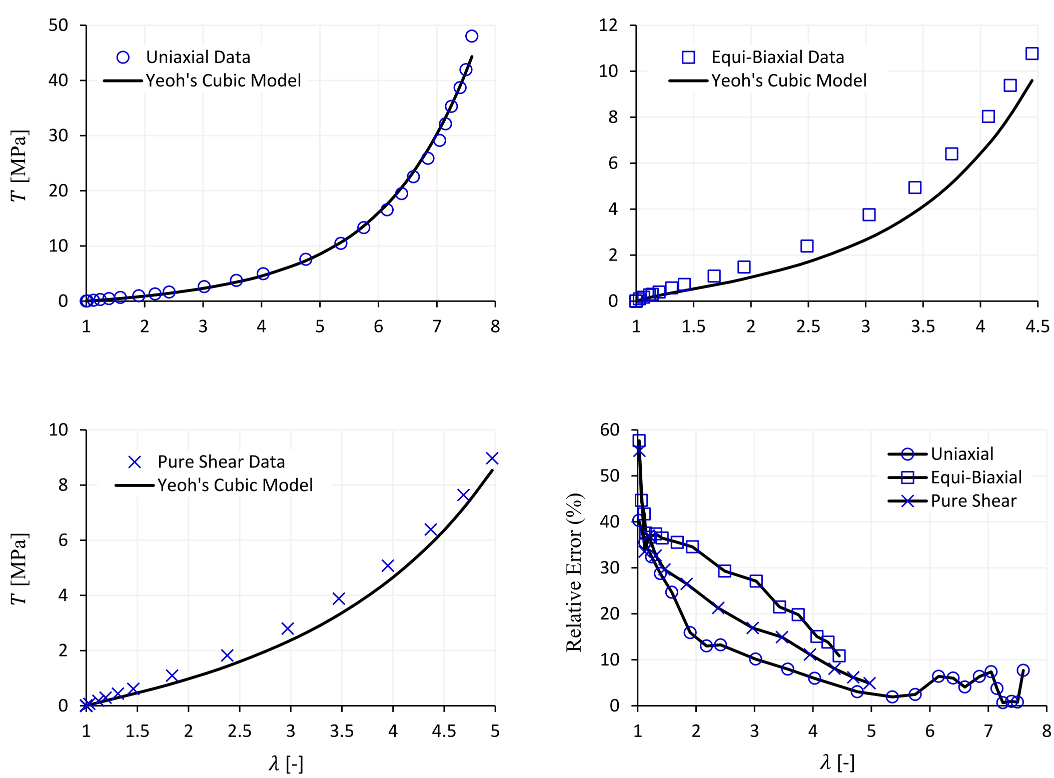

Fig. 3. Modelling results for the Treloar (1944) dataset using the Yeoh (1993) cubic model. The best fits were obtained using the simultaneous fitting procedure described in Section 3. The obtained model parameters are: $\mathscr{C}_1 = 0.13$ MPa, $\mathscr{C}_2 = 0.009$ Pa and $\mathscr{C}_3 = 28.09$ Pa.

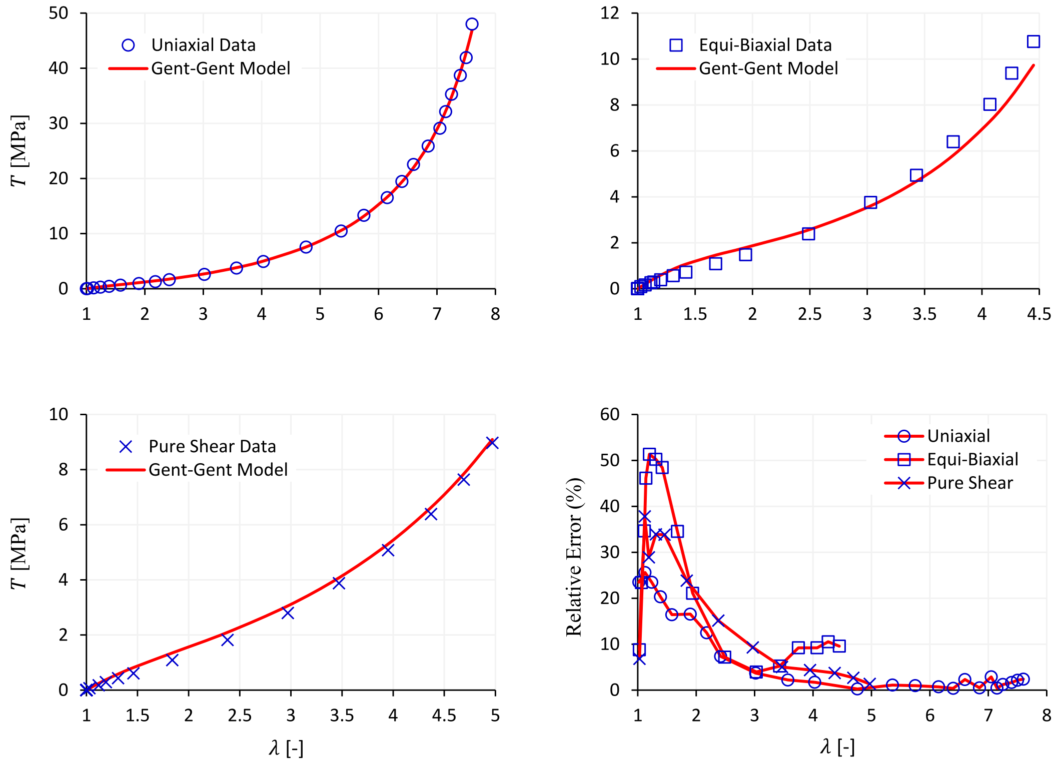

Fig. 4. Modelling results for the Treloar (1944) dataset using the Gent-Gent (Pucci and Saccomandi, 2002) model. The best fits were obtained using the simultaneous fitting procedure described in Section 3. The obtained model parameters are: $\mu_0 = 0.24$ MPa, $J_m = 77.79$ [-] and $C_2 = 0.46$ MPa.

and pure shear datasets. The same trend was also observed in relation to Kawabata et al. (1981) data (not reported here). It therefore appears that the inclusion of an I_2 term *does not* axiomatically lead to a significant improvement in modelling the mechanical behaviour of natural rubber samples compared with the proposed model. The same trend, and to a noticeably higher degree, will be also demonstrated in the next section for the case of filled rubbers.

3.2. Filled rubber

Filled rubbers exhibit a 'distinctly different' stress-strain curve than that of the unfilled rubbers (Kaliske and Heinrich, 1999), and the presence of filler additives 'distorts' the mechanical response of the host rubber compound (Dal et al., 2021). This distinction and distortion usually reflects itself via a softening effect in the stress-deformation curves, where a downward concavity is observed; see, e.g., the datasets on (carbon) filled rubber by Lahellec et al. (2004). This type of mechanical behaviour, and in particular the dataset of Lahellec et al. (2004) which pertains to a commercial rubber material developed by the Michelin group, has long been identified to be challenging for the existing hyperelastic models to capture; see, e.g., the contribution by Lopez-Pamies (2010). Indeed, to the best knowledge of the authors, the model by Lopez-Pamies (2010) may be currently one of the very few models that can successfully capture such a behaviour. This model, which has the form $W(I_1) = \sum_r \frac{3^{1-\alpha_r}}{2\alpha_r}\mu_r\left(I_1^{\alpha_r} - 3^{\alpha_r}\right)$ where $r = 1, 2, \ldots,$ and μ_r and α_r are real-valued constants, is however a purely phenomenological model and for some datasets requires at least two terms, i.e., four parameters, to capture this behaviour (Lopez-Pamies, 2010). It may therefore prove advantageous if a structurally-motivated model with fewer model parameters could be devised that is equally capable of capturing the mechanical behaviour of filled rubbers.

To this end, therefore, we consider the dataset of Lahellec et al. (2004) by way of an example to demonstrate the capability of the proposed model (1) in capturing the mechanical behaviour of filled rubbers and the inherent downward concavity of their ensuing stress-strain curves. The dataset of Lahellec et al. (2004) contains data on uniaxial and simple shear deformations of the (carbon) filled rubber specimen, which we simultaneously fit to the model using Eqs. (21) and (24). The tabulated experimental data points have been presented in Appendix A. The modelling results are shown in Fig. 5, and Table 3 lists the obtained model parameter values. Note that the parameter N is henceforth treated as a phenomenological parameter with *a priori* unrestricted values. From the plots in Fig. 5 it may be observed that the proposed model provides favourable fits to this challenging dataset, with relative errors typically below 6%.

As with the modelling results for the natural rubber specimens, we now provide a comparison with the fits provided by the other considered three-parameter models. For the sake of thoroughness, in addition to the Yeoh (1993) cubic model and the Gent-Gent model (Pucci and Saccomandi, 2002), here we also consider another three-parameter model

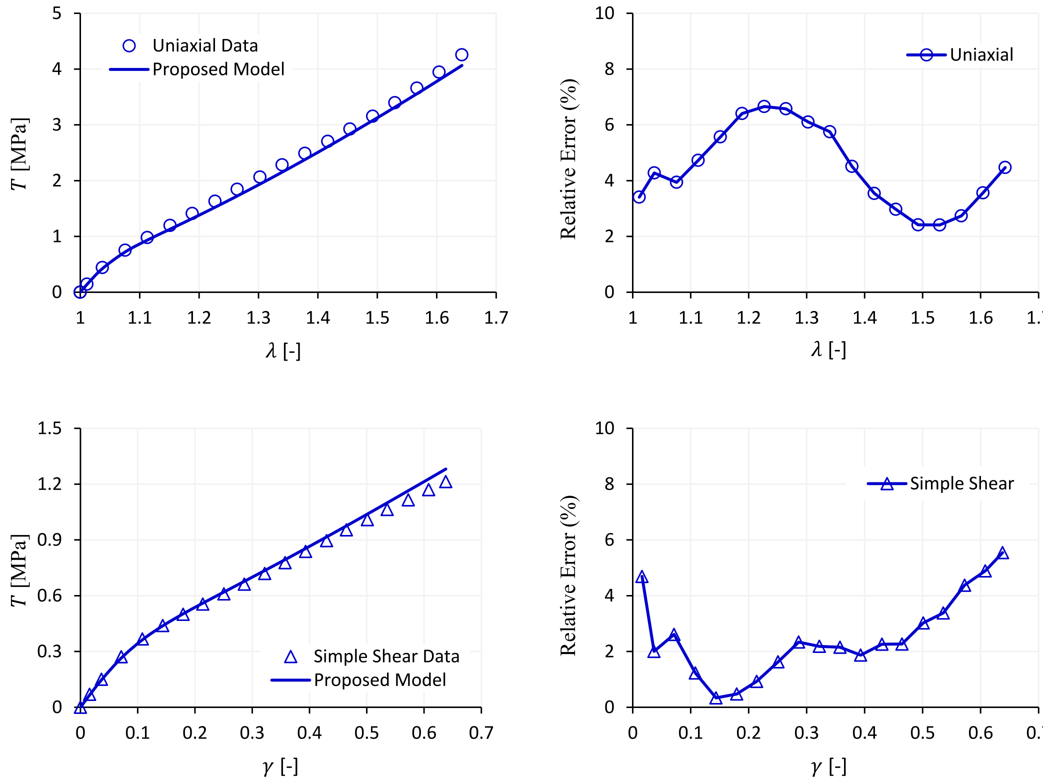

Fig. 5. Modelling results for the Lahellec et al. (2004) dataset using the proposed model (1).

Table 3
Model parameters for the Lahellec et al. (2004) dataset using the model (1).

Deformation mode	μ [MPa]	N [-]	n [-]	R^2	μ_0 [MPa][a]
Uniaxial	1.88	0.99	0.99	0.99	4.13
Simple shear				0.99	

[a] Calculated on using Eq. (3) with the given values of μ, N and n.

which too contains an I_2 term, namely the model by Carroll (2011) with the form:

$$W(I_1, I_2) = AI_1 + BI_1^4 + CI_2^{0.5}, \tag{27}$$

where A, B and C are positive model constants. The results of the simultaneous fitting of each of these models to the Lahellec et al. (2004) dataset are presented in Fig. 6. The plots in top, middle and bottom panels demonstrate the provided fits by the cubic, Gent-Gent and Carroll models, respectively.

It is readily observed that the three foregoing models all fail to provide an acceptable fit (note that the relative error plots have not been shown as the poor quality of the fits is evident). It is therefore clear that the proposed model in Eq. (1) not only provides favourable fits to the deformation of (vulcanised) natural rubber specimens, but also to filled rubbers, and to this end demonstrates *robustness* for application to modelling the mechanical behaviour of *various* rubber-like materials. The other three-parameter models considered, including the ones with an I_2 term, fail to provide such robustness.

3.3. Polymeric and silicone specimens

Nunes and co-workers have shown that the aforementioned 'softening' effects, or equivalently the downward concavity in the stress-strain curves, are not limited to filled rubbers and are also observed in the mechanical behaviour of polymeric/silicone samples (e.g., see, Nunes, 2011, Nunes and Moreira, 2013, Moreira and Nunes, 2013 and Araújo and Nunes, 2020). Therefore, as another example of application to various types of elastomers, here we consider the application of model (1) to a reported dataset pertaining to a polymeric specimen, silane modified polymer (Flextec®FT 101), due to Nunes and Moreira (2013) and Moreira and Nunes (2013). The former dataset contains data related to the simple tension and shear, and the latter study contains datasets on pure and simple shearing deformations of the specimens. The tabulated experimental data points reported in/extracted from these studies have been provided in our previous work (Anssari-Benam and Horgan, 2021).

Starting with the former study, modelling results for the dataset of Nunes and Moreira (2013) are presented in Fig. 7, while Table 4 lists the obtained model parameters. The results have been achieved by simultaneous fitting of Eqs. (21) and (24) to the data.

Subsequently we now present the modelling results pertaining to

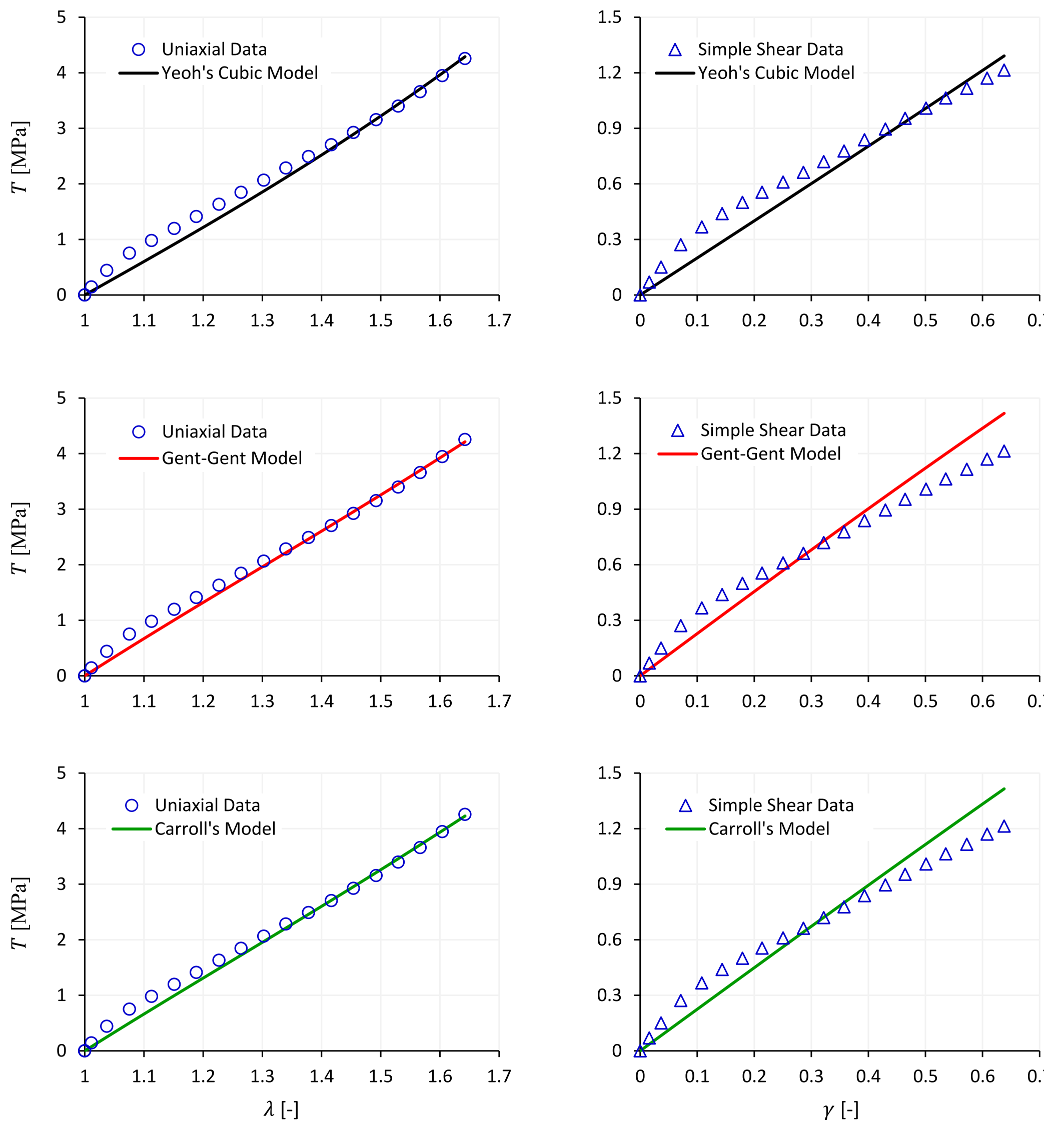

Fig. 6. Modelling results for the Lahellec et al. (2004) dataset using the cubic (top panel), Gent-Gent (middle panel) and the Carroll (bottom panel) models. The best fits were obtained using the simultaneous fitting procedure described in Section 3. The obtained model parameters are $\mathscr{C}_1 = 1.00$ MPa, $\mathscr{C}_2 = 0.015$ MPa and $\mathscr{C}_3 = 0$ Pa for the cubic model, $\mu_0 = 1.72$ MPa, $J_m = 99.89$ [-] and $C_2 = 0.84$ MPa for the Gent-Gent model, and $A = 0.87$ MPa, $B = 0.27$ Pa and $C = 0.87$ MPa for the Carroll model.

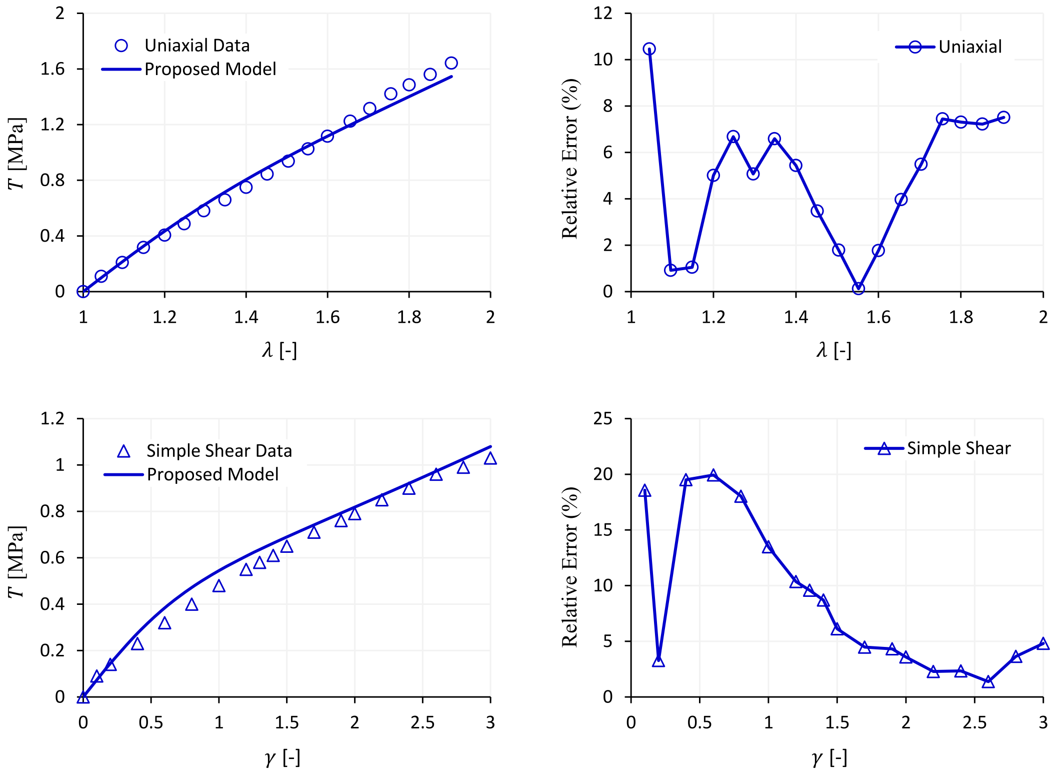

Fig. 7. Modelling results for the Nunes and Moreira (2013) dataset using the proposed model (1).

Table 4
Model parameters for the Nunes and Moreira (2013) dataset using the model (1).

Deformation mode	μ [MPa]	N [-]	n [-]	R^2	μ_0 [MPa][a]
Uniaxial	0.01	0.59	0.03	0.99	0.74
Simple shear				0.99	

[a] Calculated on using Eq. (3) with the given values of μ, N and n.

Table 5
Model parameters for the Moreira and Nunes (2013) dataset using the model (1).

Deformation mode	μ [MPa]	N [-]	n [-]	R^2	μ_0 [MPa][a]
Pure shear	0.71	−2.74	5.00	0.99	0.56
Simple shear				0.99	

[a] Calculated on using Eq. (3) with the given values of μ, N and n.

simultaneous fitting of the model to pure and simple shearing deformation datasets, due to Moreira and Nunes (2013), via Eqs. (23) and (24). Table 5 lists the obtained model parameter values, and Fig. 8 illustrates the fitting results.

We note from the values in Tables 4 and 5 that while the specimens used by Nunes and Moreira (2013) and Moreira and Nunes (2013) are of the same material (silane modified polymer), the model parameters do not possess the same values. This is likely due to the differences in the methods of preparation of the samples and the experimental setups required to do the different tests in the studies. We note, for example, that the $T-\gamma$ curves reported in the two studies, and reproduced in Figs. 7 and 8, are not the same either, and thus one would not expect of the model to return the same parameter values.

Similar to the results for the filled rubber specimen in the preceding Section, from Figs. 7 and 8 it is observed that our proposed model in Eq. (1) provides favourable fits to the deformation of the considered polymeric specimens. The other three-parameter models considered, namely the cubic, Gent-Gent and Carroll models, all fail to provide an acceptable fit and effectively produce a similar trend of fitting as with the filled rubber specimen (Fig. 6). We therefore refrain from reproducing those results here in the interest of brevity.

3.4. Soft tissue specimens: brain tissue

Finally, we consider here the application of the proposed model to the biomechanics of soft tissues, by way of providing the modelling results for a dataset containing tension, compression and simple shear of human brain (cortex) tissue. In a comprehensive study of the mechanical behaviour of the brain tissue, Budday et al. (2020b) performed uniaxial and simple shear tests on human brain tissue samples, and used the (one-term) Ogden model to model the ensuing observed mechanical behaviour of the specimens. Brain tissue is considered to be extremely soft, and the Ogden model appears to be the most commonly used model for capturing the biomechanics of the brain tissue. However, as was shown by Anssari-Benam and Horgan (2021), the one-term Ogden model so calibrated with the data will be non-convex. Therefore, while the fitting result may be better than that provided by most other existing models, it is not mechanically consistent. Motivated by this finding, here we present the results of the application of the model in Eq. (1) to the aforementioned dataset and show that the model provides comparable fitting quality to the one-term Ogden model, with even lower relative errors, while remaining convex *a priori*. The lower quality of the fits provided by the three-parameter cubic, Gent-Gent and Carroll models will also be demonstrated.

Accordingly, Fig. 9 presents the modelling results via the simultaneous fitting of Eqs. (21) and (24) to the Budday et al. (2020b) dataset. The bottom panel shows the iso-energy plots of the strain energy function W, demonstrating the convexity of the proposed function over the domain of the deformation. Table 6 also includes the obtained model parameter values. We note from the value of N in Table 6 that $N > 1$ so that in this application the model (1) is being tested within its original

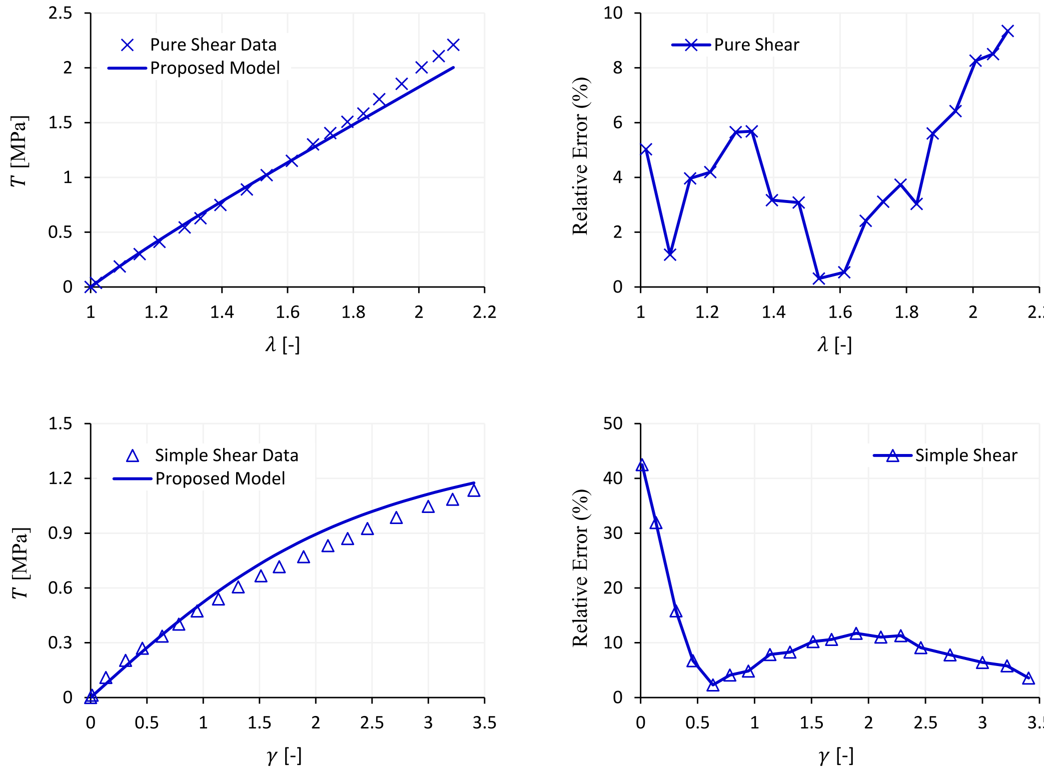

Fig. 8. Modelling results for the Moreira and Nunes (2013) dataset using the proposed model (1).

structural framework. The tabulated experimental data points extracted from the study of Budday et al. (2020b) have been presented in our previous work (Anssari-Benam and Horgan, 2021).

While at the first glance it may be observed that the fits have some limitations, we also present here the modelling results using the one-term Ogden model, as reported in Budday et al. (2020b). Recall that the one-term Ogden model has the form $W_{Ogden\ 1} = \frac{2\mu}{\alpha^2}\left(\lambda_1^\alpha + \lambda_2^\alpha + \lambda_3^\alpha - 3\right)$, where μ and α are the two model parameters. The results are shown in Fig. 10.

Comparing the results in Figs. 9 and 10 indicates that while the Ogden model better captures the asymmetry of the compression-tension data, it produces higher maximum errors than the proposed model in Eq. (1). However, and more importantly, the proposed model remains convex, while the one-term Ogden model produces non-convex results. Recall that subject to the constraint (7), the logarithmic term is always well-defined and convex, which ensures that the W function remains convex *a priori*. It therefore appears that the proposed model (1) offers clear advantages over the one-term Ogden model for application to the brain tissue biomechanics, albeit with an additional model parameter.

Therefore, taking all the datasets and fitting results presented in Section 3 into account, the proposed model in Eq. (1) appears to provide a reliable degree of *robustness*, with the capability of capturing the mechanical behaviour of a wide range of rubber-like materials including unfilled and filled rubbers, polymers and (extremely) soft tissues. The existing three-parameter models in the literature considered here failed to show this robustness, and typically resulted in larger errors compared with the proposed model.

4. Concluding remarks

With the broader aim of working towards the development of a class of *comprehensive* hyperelastic models, a three-parameter strain energy function was utilised in this study for application to *various types* of isotropic rubber-like materials. The response function for this model was first devised by Anssari-Benam (2021) as a [1/1] Padé approximant of the inverse Langevin function $\mathscr{L}^{-1}$. Here, on using the constraints (2) and (7), we generalised the application of the model to unfilled and filled rubbers, polymers and soft tissues. It was shown here that this generalised neo-Hookean strain energy function, in contrast to many existing models of this kind in the literature and to those which also embody an I_2 term, provides a reliable *robustness* for application to different types of soft solids. In addition, it was also demonstrated that the proposed model is the *parent* model to some of the existing limiting chain extensibility models in the literature such as the celebrated Gent (1996) model and the model in Anssari-Benam and Bucchi (2018, 2021). With only three constitutive parameters, finding an optimal fit using this model circumvents the uniqueness issues identified by Ogden et al. (2004) for models with higher number of parameters. Marckmann and Verron (2006) concluded in their review that "… models with only two or three material parameters are unable to predict the whole range of strain …". Our results here using the proposed model dispel this pessimistic notion. The structural roots of this model also provides a degree of physical objectivity and mathematical underpinning for the model parameters, not enjoyed by many of the existing models.

We note that some studies, through experimental observations (see, e.g., Horgan and Saccomandi, 1999 and Destrade et al., 2015), or deriving universal relationships (e.g., Saccomandi, 2001; Wineman, 2005; Horgan and Smayda, 2012) have highlighted the necessity of inclusion of an I_2 term in the functional form of the strain energy function. Our results here indicate that even with an I_2 term, there is no axiomatic guarantee that a model will be able to provide universality in capturing the mechanical behaviour of various rubber-like materials. By contrast, the functional form of the proposed model in Eq. (1) appears to be *rich* enough to provide this generality, at least within the bounds of the type of elastomers considered in this study. Nevertheless, in applications where the inclusion of an I_2 term becomes necessary, the proposed model herein may be easily supplemented with such terms. Indeed, for the special case of the model where $n = 3$, i.e., the model used by Anssari-Benam and Bucchi (2021), this extension has been applied in our previous studies and the favourable comparative modelling results

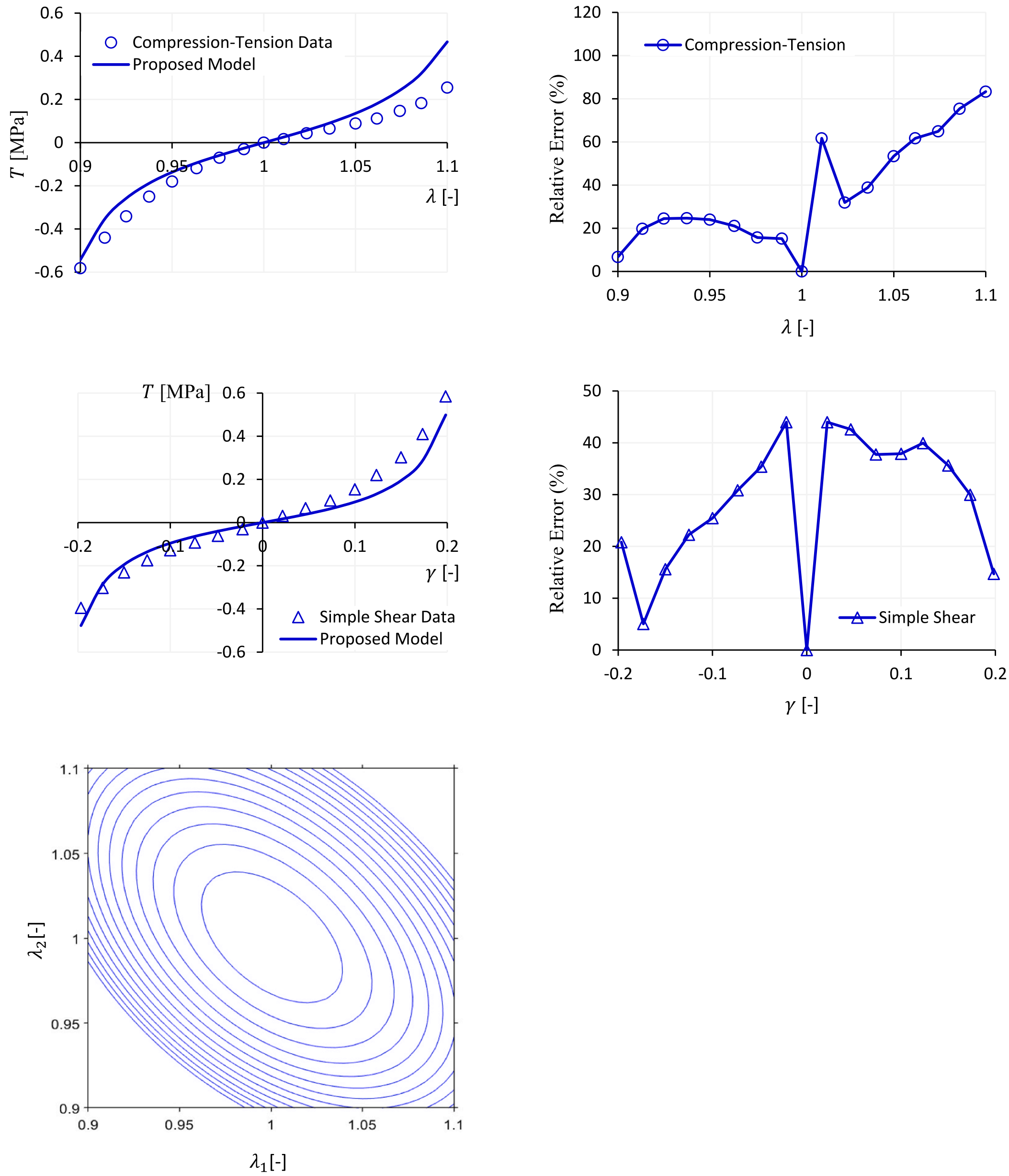

Fig. 9. Modelling results for the Budday et al. (2020b) brain tissue dataset using the proposed model (1). The bottom panel shows the iso-energy plots of the strain energy function W in the plane of the principal stretches (λ_1, λ_2), demonstrating the convexity of the model.

Table 6
Model parameters for the Budday et al. (2020b) brain tissue dataset using the model (1).

Deformation mode	μ [kPa]	N [-]	n [-]	R^2	μ_0 [kPa][a]
Compression-tension	0.016	1.02	14.04	0.95	0.79
Simple shear				0.97	

[a] Calculated on using Eq. (3) with the given values of μ, N and n.

have been presented (see Anssari-Benam et al., 2021a, 2021c). The advantage of having only three parameters in the proposed model is that even if an I_2 term was added, the overall number of model parameters would be four, which is still in the lower end of the number of parameters contained in models ranked as the 'best' in recent reviews of Marckmann and Verron (2006) and Dal et al. (2021).

Of particular note is the success of the proposed model in capturing the typical 'softening' effect, or the downward concavity, in the stress-strain curves of filled rubbers. Up to now, to the best of our knowledge, the only successful model in this regard has been that of Lopez-Pamies (2010), also echoed in the work of, for example, Nunes and Moreira (2013) in relation to polymers. However, with three constitutive parameters and a structural basis, our proposed model in Eq. (1) provided favourable fits to the filled rubber and polymer datasets. In a previous work (Anssari-Benam and Horgan, 2021), we presented a generalisation of the two-parameter model of Anssari-Benam and Bucchi (2021) for application to the shear deformation of these specimens. That generalisation resulted in improved fits compared with the original model or that of the Gent model. However, the three-parameter model proposed here provides much improved fits, and with simultaneous fitting to other deformation datasets of the specimens.

The four-parameter extended tube model of Kaliske and Heinrich (1999) was identified by Marckmann and Verron (2006) as the best

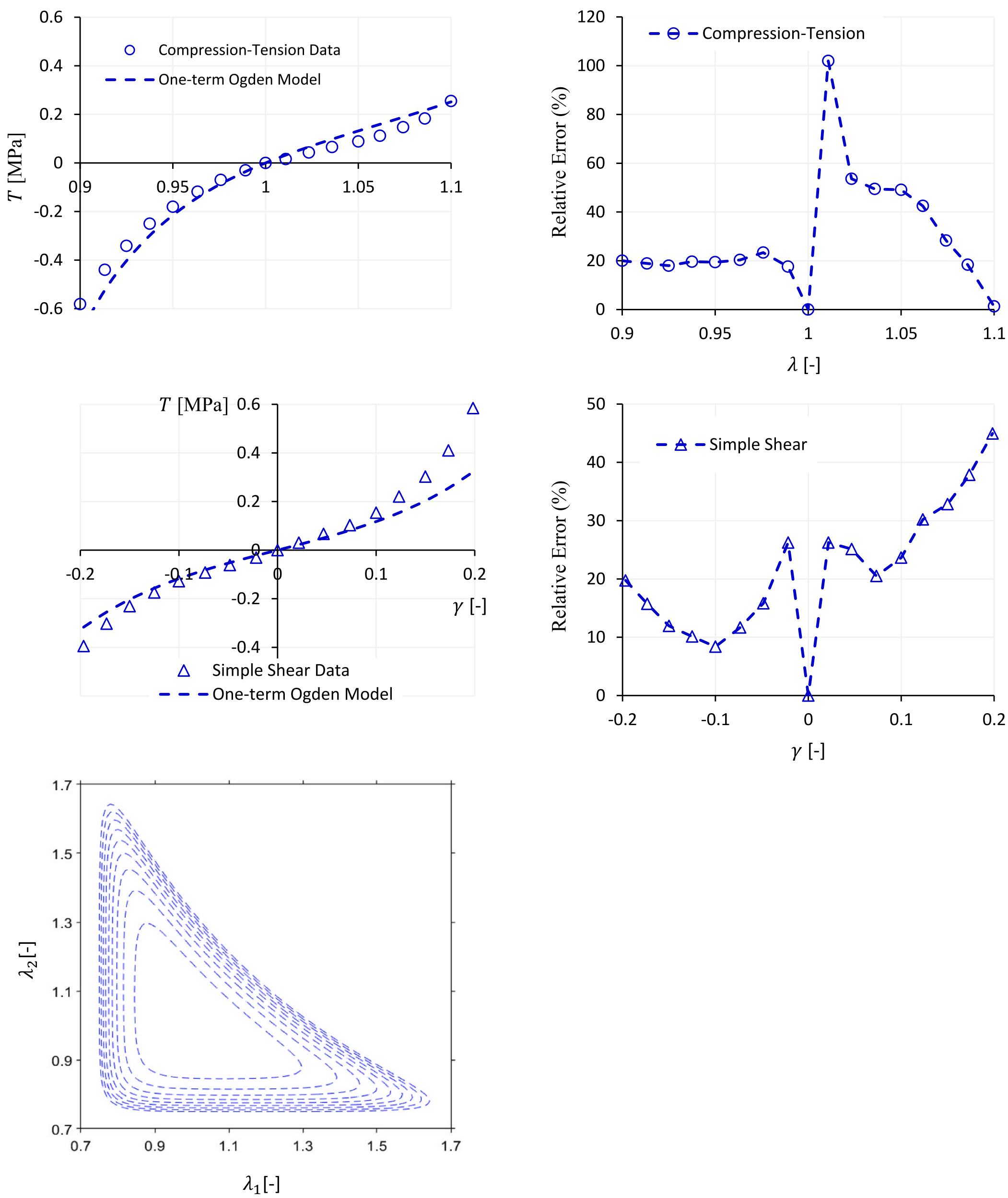

Fig. 10. Modelling results for the Budday et al. (2020b) brain tissue dataset using the one-term Ogden model. The optimal fit was found by Budday et al. (2020b) with $\mu = 1.04$ kPa and $\alpha = -17.33$ [-]. The bottom panel shows the iso-energy plots which demonstrates that the one-term Ogden model is *not* convex for this fitting.

available model, while the review of Dal et al. (2021) recommends the five-parameter model by Miehe et al. (2004). The performance of both these models in relation to filled rubber samples or soft tissues remains untested, as the foregoing reviews mainly considered the datasets of Treloar (1944) and Kawabata et al. (1981) for the performance assessment of the models. Therefore, the degree of universality offered by these two models in capturing the mechanical behaviour of various rubber-like materials is still an open question. Moreover, the elaborate mathematical form of these models combined with the extra number of model parameters that they contain does not necessarily offer them as more attractive candidates compared with the proposed model here. The encouraging results obtained in this study perhaps warrant the application of the proposed three-parameter model to areas where the aforementioned two models have been employed.

Another area of development with a promising potential is the application of the proposed model to the biomechanics of soft tissues. In its current form, the proposed model may serve to provide a more accurate representation of the mechanical behaviour of the *isotropic matrix*. The popular neo-Hookean model often employed in the literature to this effect is known to *not* always provide an accurate description of the elastin network deformation (see, e.g., Anssari-Benam and Bucchi, 2018). Collagenous soft tissues exhibit a strong anisotropy, and a next step is therefore to extend the proposed model to include such effects. For the Anssari-Benam and Bucchi (2021) subset model, this specialisation has been developed and presented (Anssari-Benam et al., 2020), which may pave the way for the generalisation of the proposed model herein too.

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.

Appendix A. Tabulated experimental data due to Lahellec et al. (2004)

Table A1
Lahellec et al. (2004) data as reported in Lopez-Pamies (2010).

λ [−]	T_{uni} [MPa]	γ [−]	T_{ss} [MPa]
1	0	0	0
1.01	0.14	0.02	0.07
1.04	0.44	0.04	0.15
1.07_5	0.75	0.07	0.27
1.11	0.98	0.11	0.37
1.15	1.20	0.14	0.44
1.19	1.41	0.18	0.50
1.23	1.63	0.21	0.55
1.26	1.85	0.25	0.61
1.30	2.06	0.29	0.66
1.34	2.28	0.32	0.72
1.38	2.49	0.36	0.78
1.42	2.70	0.39	0.84
1.45	2.92	0.43	0.90
1.49	3.15	0.46	0.95
1.53	3.40	0.50	1.01
1.57	3.66	0.53_5	1.06
1.60	3.95	0.57	1.11_5
1.64	4.25_5	0.61	1.17
		0.64	1.21

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