**A generalised neo-Hookean strain energy function for application to the finite
deformation of elastomers**

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Abstract

We present a new model within the class of generalised neo-Hookean strain energy functions for application to the finite deformation of incompressible elastomers. The model has a simple form with only two parameters, namely μ and N with structural roots, and is derived within the classical framework of statistical mechanics for freely jointed molecular chains in rubber elasticity. Using existing experimental datasets of uniaxial, biaxial, and pure shear deformations, including Treloar's canonical data, we show that the proposed model captures the deformation behaviours of each elastomer specimen with a single set of μ and N values (with only a narrow variation range). We conduct qualitative and quantitative comparative analyses to highlight the advantages of the proposed model, and demonstrate improved fits versus the celebrated Gent model. Given its structural molecular basis, simplicity, small number of parameters, and its capability to describe many deformation modes of a specimen using a single set of parameter values, we propose this model for application to finite deformation of elastomers.

Keywords: Strain energy function, hyperelasticity, elastomers, molecular chains, finite deformation.

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1. Introduction

The aim of this manuscript is to propose the following generalised neo-Hookean strain energy function for application to modelling the large deformation of incompressible elastomers:

$$W = \mu N \left[\frac{1}{6N} (I_1 - 3) - \ln \left(\frac{I_1 - 3N}{3 - 3N} \right) \right], \quad (1)$$

where μ and N are structural model parameters that will be defined in §2, and I_1 is the first principal invariant of the right Cauchy-Green tensor.

The background for devising this model stemmed from a soft tissue biomechanics perspective, where the authors were trying to find a suitable strain energy function that could better describe the large deformation endured by the ‘isotropic matrix’ in aortic valve tissue specimens. The prevalent approach in continuum-based modelling of soft tissue deformations, following the seminal work of Holzapfel et al. [1], is to use an additive split of the total strain energy function in the form of $W_{iso} + W_{aniso}$, to account for the contribution of the isotropic matrix and the anisotropy endowed by the fibres, respectively. In this regard, it is a common practice to use the well-known neo-Hookean strain energy function as W_{iso} . In the case of our samples, however, the neo-Hookean function proved to be wholly inadequate, leading us instead to utilise an exponential strain energy function of the form: $\alpha \exp[\beta(I_1 - 3)]$, with α and β being phenomenological constants (see [2] for more details). To rationalise a less phenomenological and more structurally relevant model, based on the framework of statistical mechanics originally applied in rubber elasticity, we customised a new strain energy function similar to that in Eq. (1), which successfully captured the large deformation behaviour of the elastin network [3] and the isotropic matrix of the aortic valve [4].

Given the traditional application of the generalised neo-Hookean strain energy functions in rubber elasticity and the common root from which those functions and our model in Eq. (1) are derived, i.e. the molecular theory of rubbers, it is our aim herein to propose and demonstrate the application of our model to the finite deformation of elastomers and rubber-like materials. In addition, following expert points highlighted by Puglisi and Saccomandi [5] and Destrade

et al. [6] as regards the shortcomings of the existing models, we note the advantages that our model may offer over the existing models of the same class in the literature.

First, a model should ideally include only a small number of parameters. Embodying a large number of model parameters gives rise to problems surrounding implementation and optimisation, resulting in the existence of a number of ‘optimal’ parameter sets and uniqueness issues. A systematic analysis on this point is presented by Ogden et al. [7] where the sensitivity of the fitting procedure and the possibility of finding a large number of optimal sets of parameters have been highlighted for the third- and fourth-order Ogden model. While the Ogden model does not belong to the class of generalised neo-Hookean functions, a similar sentiment may however be extended to models that incorporate an excessive number of model parameters.

Second, model parameters should ideally have physical meaning and therefore be objective. A model with a sound structural basis will curb the need for phenomenological modifications. For example, the cubic function of $(I_1 - 3)$ presented by Yeoh [8] or the linear summation of $(I_1^{\alpha_i} - 3^{\alpha_i})$ considered by Lopez-Pamies [9], while providing satisfactory fits to the considered deformation datasets, were originally proposed as phenomenological modifications to Mooney-Rivlin and neo-Hookean models, respectively. It is worth noting that while for particular values of material constants phenomenological models may reduce to structurally-derived models, a general physical interpretation of the phenomenological parameters in such models is difficult to underpin. Models with direct structural basis will relieve the pressure on efforts seeking to reconcile the molecular and continuum/phenomenological approaches in rubber elasticity, which appears to form a notable volume of the literature. An explicit account of these efforts can be seen in [10], with more recent contributions in [11] and [12] for providing a mathematical underpinning for the molecular basis of the Gent model [13], which was originally proposed as a phenomenological model.

Third, a constitutive model must be able to satisfactorily capture many deformation modes of a specimen while providing universality. That is, ideally, a single set of model parameters should capture many deformation modes of the same material, and all modes of deformation should ideally be captured by the model. For example, the neo-Hookean function only includes one material parameter, namely the infinitesimal shear modulus μ_0 ; however, its shortcomings

to capture the whole-range behaviour of elastomers, particularly at higher deformations, have been well documented. Many datasets may also be found in the literature which celebrated models such as the Gent and Arruda-Boyce fail to adequately address (e.g., see [9] for more details). The current literature suggests a general lack of existence of a universal model for rubberlike materials [6]. Universality in this context is not taken to mean a model that can capture the behaviour of all elastomeric materials, but different deformation modes for the same specimen. Examples of this shortcoming have been noted and discussed in, e.g., [14].

At this juncture, however, it may be worthwhile to recall that a ‘universal’ model ought to not only incorporate I_1 , but also I_2 , the second principal invariant of the right Cauchy-Green tensor. Specifically, any valid strain energy function W must at least be consistent with the requirements of fourth-order weak nonlinear elasticity, which in turn necessitates the existence of three constitutive parameters in the model function [15,16]. In view of the classical models of rubber elasticity, this requirement may be deemed tantamount to inclusion of an I_2 term (accompanied by a C_2 parameter) in the model. In addition, via deriving universal relationships, the *a priori* dependence of the strain energy function W on I_2 in order to appropriately capture certain deformation modes such as simple shear, torsion and non-homogenous deformations has been established (see, e.g., [17-19] amongst others). Therefore, a valid constitutive model has to include I_2 terms. Clearly, generalised neo-Hookean strain energy functions, by definition, preclude I_2 terms. Moreover, the origin of the C_2 term in rubber elasticity is related to the non-affine deformation of the entanglement network formed by physical and chemical crosslinks between molecular chains [20], which is not taken into account in the classical approach of average chain models when deriving the mathematical form of the generalised neo-Hookean functions. Hence, this class of strain energy functions may be deemed as a subset of what may be a universally valid model, whilst providing mathematical simplicity and a satisfactory characterisation of many experimental datasets.

Within the bounds of a generalised neo-Hookean strain energy function, our proposed model in Eq. (1) embodies only two material parameters, has a direct structural root based on the statistical mechanics framework for molecular chains in rubber elasticity, and as will be shown in proceeding sections using published data, is capable of describing many deformation modes of the same rubber specimens with a single set of parameter values obtained by simultaneous fitting to the experimental data. The derivation of this model is presented in §2, followed by

the application of the model to extant experimental data in §3, where we also compare the fitting results with the Gent model. In §4 a comparative analysis between our proposed model and some of the well-established models of the same class in the literature is provided, and concluding remarks are conferred in §5. We thereby present our model as an attractive choice of incompressible isotropic hyperelastic strain energy function next to the existing models in the literature.

2. Theoretical background

2.1. Model development

The derivation of the general form of the strain energy function of a network of freely jointed molecular chains based on the principles of statistical mechanics and using the (inverse) Langevin function, first introduced by James and Guth [19] for application to large deformations in rubber elasticity, has been presented elsewhere (see, e.g., [22,23]). Here we provide a concise background for the sake of completeness and present our customisation for arriving at the particular form of the proposed strain energy function W in Eq. (1).

For lucidity in the idealisation of the kinematics of deformation, let us consider the unit cube cell shown in Fig. 1 such that molecular chains form its spatial diagonals (Fig. 1a). Employing Boltzmann's equation, it is straightforward to show that the free energy of a single chain ψ may be given by the following function (see, e.g., [3]):

$$\psi = NkT \left[\frac{\lambda_c}{\sqrt{N}} \ell^{-1} \left(\frac{\lambda_c}{\sqrt{N}} \right) + \ln \frac{\ell^{-1} \left(\frac{\lambda_c}{\sqrt{N}} \right)}{\sinh \ell^{-1} \left(\frac{\lambda_c}{\sqrt{N}} \right)} \right] + \psi_0, \quad (2)$$

where ψ_0 is a constant such that the free energy of the chain ψ would be zero in the undeformed configuration, k is the Boltzmann constant, T is the absolute temperature, N is the number of Kuhn segments of a chain, λ_c is the chain stretch and ℓ^{-1} is the inverse of the Langevin function ℓ defined as $\ell(\beta) = \coth \beta - \frac{1}{\beta}$. Note that in the statistical treatment of a single chain, the geometry of the chain is idealised as a composition of N segments of equal length l , referred to as Kuhn segment length, as shown in Fig 1b.

Fig. 1 - (a) an idealised molecule network structure in a representative cube cell; (b) geometry of a single chain in the 3D cube.

If the end-to-end distance of a chain in its undeformed and deformed configurations are denoted by r_0 and r , respectively, then the chain stretch λ_c is $\lambda_c = \frac{r}{r_0}$. For the considered single chain in the undeformed network shown in Fig. 1b, the unstrained end-to-end distance of the chain r_0 is: $r_0 = a_0\sqrt{3}$, where a_0 is the length of the undeformed sides of the cube. However, to link the macro-level (cube cell) to the micro-level (single chain) deformation, we assume an *affine* relationship between the macro stretch λ and the chain stretch λ_c , consistent with the bulk of the literature pertaining to *generalised neo-Hookean* strain energy functions [24]. Accordingly, after deformation, the end-to-end chain vector will become: $\mathbf{r} = a_0(\lambda_1\mathbf{e}_1 + \lambda_2\mathbf{e}_2 + \lambda_3\mathbf{e}_3)$, where λ_1 , λ_2 and λ_3 are the principal (macro) stretches. Hence, the magnitude of $\mathbf{r}$ will be $r = a_0\sqrt{\lambda_1^2 + \lambda_2^2 + \lambda_3^2} = a_0\sqrt{I_1}$, where I_1 is the first principal invariant of the right Cauchy-Green deformation tensor $\mathbf{C}$ and is given by $I_1 = \lambda_1^2 + \lambda_2^2 + \lambda_3^2$. It may now be readily observed that $\lambda_c = \sqrt{\frac{I_1}{3}}$.

Assuming that the network is comprised of n chains of equal Kuhn segment length l per unit volume and all the chains stretch uniformly, the free energy of the full network $\psi_{network}$ may

be obtained as $\psi_{network} = \sum_{i=1}^n \psi^i$ [21]. Noting that the energy function W for the macro-level deformation of the continuum is $W = \psi_{network} \left(\sqrt{\frac{I_1}{3}} \right)$ [24], the strain energy function of the continuum W may be explicitly derived from Eq. (2) as:

$$W = nkTN \left[\sqrt{\frac{I_1}{3N}} \ell^{-1} \left(\sqrt{\frac{I_1}{3N}} \right) + \ln \frac{\ell^{-1} \left(\sqrt{\frac{I_1}{3N}} \right)}{\sinh \ell^{-1} \left(\sqrt{\frac{I_1}{3N}} \right)} \right] + \psi_0. \quad (3)$$

Again, note that ψ_0 is a constant so that W would be zero in the undeformed state. This equation is similar to that of the strain energy function in Arruda and Boyce [22], equation (7) therein, while a similar relationship has also been demonstrated for a full network model by Beatty [23].

We now, however, present our customisation of W . Establishing the partial derivative of W in Eq. (3) with respect to I_1 we find:

$$\begin{aligned} \frac{\partial W}{\partial I_1} = & nkTN \ell^{-1} \left(\sqrt{\frac{I_1}{3N}} \right) \frac{\partial}{\partial I_1} \sqrt{\frac{I_1}{3N}} + nkTN \sqrt{\frac{I_1}{3N}} \frac{\partial}{\partial I_1} \ell^{-1} \left(\sqrt{\frac{I_1}{3N}} \right) \\ & + nkTN \frac{\partial}{\partial I_1} \ln \frac{\ell^{-1} \left(\sqrt{\frac{I_1}{3N}} \right)}{\sinh \ell^{-1} \left(\sqrt{\frac{I_1}{3N}} \right)}. \end{aligned} \quad (4)$$

Using the change of variable $\ell^{-1} \left(\sqrt{\frac{I_1}{3N}} \right) = \xi$, the above equation simplifies to:

$$\frac{\partial W}{\partial I_1} = nkTN \xi \frac{\partial}{\partial I_1} \sqrt{\frac{I_1}{3N}} + nkTN \sqrt{\frac{I_1}{3N}} \frac{\partial \xi}{\partial I_1} + nkTN \frac{\partial}{\partial I_1} \ln \frac{\xi}{\sinh \xi}. \quad (5)$$

However, in the above equation we note that:

$$\frac{\partial}{\partial I_1} \ln \frac{\xi}{\sinh \xi} = \frac{\partial \ln \frac{\xi}{\sinh \xi}}{\partial \xi} \frac{\partial \xi}{\partial I_1} = \frac{\sinh \xi - \xi \cosh \xi}{\xi \sinh \xi} \frac{\partial \xi}{\partial I_1} = \left(\frac{1}{\xi} - \coth \xi \right) \frac{\partial \xi}{\partial I_1}, \quad (6)$$

where $\frac{1}{\xi} - \coth \xi = -\ell(\xi) = -\ell \left[\ell^{-1} \left(\sqrt{\frac{I_1}{3N}} \right) \right] = -\sqrt{\frac{I_1}{3N}}$. Therefore, Eq. (5) becomes:

$$\frac{\partial W}{\partial I_1} = nkTN\xi \frac{\partial}{\partial I_1} \sqrt{\frac{I_1}{3N}} + nkTN \sqrt{\frac{I_1}{3N}} \frac{\partial \xi}{\partial I_1} - nkTN \sqrt{\frac{I_1}{3N}} \frac{\partial \xi}{\partial I_1} = nkTN\xi \frac{\partial}{\partial I_1} \sqrt{\frac{I_1}{3N}}, \quad (7)$$

which may alternatively be re-written as:

$$\frac{\partial W}{\partial I_1} = \frac{1}{2\sqrt{3}} nkT\sqrt{N} \frac{1}{\sqrt{I_1}} \ell^{-1} \left(\sqrt{\frac{I_1}{3N}} \right). \quad (8)$$

The natural next step in establishing W is to perform integration over Eq. (8). However, the inverse Langevin function appearing in Eq. (8) is not amenable to mathematical manipulation in a straight-forward manner. The literature suggests different forms of approximating this function, often using a truncated Taylor series or a Padé approximation. We have demonstrated in a previous study that a Padé approximation may be a more accurate representation [3]. In addition, inherent to the characteristics of the inverse Langevin function is its asymptotic behaviour, leading to a singularity point. The series expansion approximation essentially eliminates this feature, while a Padé approximation preserves this inherent characteristic. Accordingly, we use a Padé approximation introduced by Cohen [25] of the form:

$$\ell^{-1} \left(\sqrt{\frac{I_1}{3N}} \right) \approx \frac{\sqrt{\frac{I_1}{3N}} \left[3 - \left(\frac{I_1}{3N} \right) \right]}{1 - \left(\frac{I_1}{3N} \right)}. \quad (9)$$

Note that the above rational approximation and the original inverse Langevin function both have the same vertical asymptote, i.e. at the same point, as highlighted by Horgan and Saccomandi [26]. The reason that preserving this point of discontinuity may be useful is that it would allow prediction of a limiting stretch, beyond which the network may not stretch. This characteristic would be an additional structural feature built in the proposed W function which will be accounted for by using a Padé approximation while it would be lost in a truncated Taylor series expansion.

Substituting from Eq. (9) into Eq. (8), we obtain for W :

$$W = \frac{1}{6} nkT [I_1 - 6N \ln(I_1 - 3N)] + w_0, \quad (10)$$

where the constant w_0 may be viewed as a similar parameter to ψ_0 in Eq. (3), i.e. w_0 is a constant so that W would be zero in the undeformed state where $I_1 = 3$; therefore:

$$w_0 = -\frac{1}{6}nkT[3 - 6N \ln(3 - 3N)]. \quad (11)$$

Substituting this expression into Eq. (10) yields the final form of our proposed W as:

$$W = \mu N \left[\frac{1}{6N} (I_1 - 3) - \ln \left(\frac{I_1 - 3N}{3 - 3N} \right) \right]. \quad (12)$$

Note that $\mu = nkT$, a customary notation in molecular chain models [26,27]. However, we emphasise that μ in our model should not be confused with the infinitesimal shear modulus μ_0 .

Indeed, on using $\mu_0 = 2 \left(\frac{\partial W}{\partial I_1} \right)_{I_1=3}$ we find that $\mu = \mu_0 \left(\frac{3-3N}{1-3N} \right)$.

2.2. Convexity of the proposed W function

As stipulated by Ball [28], in many classical applications of finite elasticity it suffices for the strain energy function W to satisfy the condition of ‘polyconvexity’. Here, however, we opt for the more stringent condition of ‘strict local convexity’ which requires that the Hessian of W is positive definite. We remind the reader of the order of strength of the conditions of convexity (see, e.g., [29]):

$$\text{Convexity} \Rightarrow \text{polyconvexity} \Rightarrow \text{quasiconvexity} \Rightarrow \text{rank one convexity}.$$

Accordingly, the Hessian matrix in relation to λ may be defined as:

$$\mathbf{H} = \begin{bmatrix} \frac{\partial^2 W}{\partial \lambda_1^2} & \frac{\partial^2 W}{\partial \lambda_1 \partial \lambda_2} \\ \frac{\partial^2 W}{\partial \lambda_2 \partial \lambda_1} & \frac{\partial^2 W}{\partial \lambda_2^2} \end{bmatrix}, \quad (13)$$

where $\mathbf{H}$ denotes the Hessian matrix. Calculating the terms $\frac{\partial^2 W}{\partial \lambda_1 \partial \lambda_2}$ and $\frac{\partial^2 W}{\partial \lambda_2 \partial \lambda_1}$ in view of Eq.

(13), it may be observed that $\frac{\partial^2 W}{\partial \lambda_1 \partial \lambda_2} = \frac{\partial^2 W}{\partial \lambda_2 \partial \lambda_1} = 4nkTN \frac{\lambda_1 \lambda_2}{(3N - I_1)^2}$, and hence $\mathbf{H}$ is symmetric.

Therefore, in order for $\mathbf{H}$ to be positive definite, it is necessary that $\frac{\partial^2 W}{\partial \lambda_1^2}$, $\frac{\partial^2 W}{\partial \lambda_2^2}$, $\det(\mathbf{H})$ and the eigenvalues of $\mathbf{H}$ are all positive. Thus:

$$\left\{ \begin{array}{l} \frac{\partial^2 W}{\partial \lambda_1^2} > 0 \Rightarrow \frac{12\lambda_1^2 N}{(3N - I_1)^2} > -\frac{9N - I_1}{3N - I_1} \\ \frac{\partial^2 W}{\partial \lambda_2^2} > 0 \Rightarrow \frac{12\lambda_2^2 N}{(3N - I_1)^2} > -\frac{9N - I_1}{3N - I_1} \\ \det \mathbf{H} > 0 \Rightarrow \left(\frac{9N - I_1}{3N - I_1} \right)^2 > -\frac{12N(9N - I_1)(\lambda_1^2 + \lambda_2^2)}{(3N - I_1)^3} \\ \text{eigenvalues}(\mathbf{H}) > 0 \Rightarrow \text{both roots of the 'characteristic equation' must be positive} \end{array} \right. \quad (14)$$

It may readily be observed that all above four inequalities are satisfied *a priori* for $N > \frac{I_1}{3}$ and $< \frac{I_1}{9}$. Therefore, the proposed W in Eq. (12) satisfies the condition of strict local convexity. Note, however, that from a mathematical point of view, the inverse Langevin function $\ell^{-1}(x)$ is only defined within the domain $-1 < x < 1$, which for the variable $\sqrt{\frac{I_1}{3N}}$ is equivalent to $-1 < \sqrt{\frac{I_1}{3N}} < 1$, requiring *a priori* $N > \frac{I_1}{3}$.

As noted in the foregoing, by satisfying the condition of strict local convexity our model axiomatically also satisfies the condition of polyconvexity. However, for the sake of completeness, we recall that the condition of polyconvexity for an incompressible generalised neo-Hookean strain energy function W requires:

$$\left\{ \begin{array}{l} \frac{dW}{dI_1} > 0, \\ \frac{dW}{dI_1} + 2I_1 \frac{d^2 W}{dI_1^2} > 0. \end{array} \right. \quad (15)$$

Noting:

$$\left\{ \begin{array}{l} \frac{dW}{dI_1} = \mu N \left(\frac{I_1 - 9N}{6NI_1 - 18N^2} \right), \\ \frac{d^2 W}{dI_1^2} = \frac{\mu N}{(I_1 - 3N)^2}, \end{array} \right. \quad (16)$$

and that by definition $I_1, N, \mu > 0$, the inequality in (15) reduces to $N > \frac{I_1}{3}$ or $N < \frac{I_1}{9}$. Since for the domain of the inverse Langevin function to be valid we require *a priori* $N > \frac{I_1}{3}$, the

condition of strict polyconvexity in inequality (15) is satisfied *a priori* for our proposed strain energy function.

3. Application to experimental data

As a means of measuring how well the proposed model captures the deformation behaviour of elastomers, we now present the application of this model to a range of previously published experimental data. We choose datasets containing uniaxial, biaxial and pure shear deformations of rubber samples including the canonical data due to Treloar [30]¹ and a particular dataset due to Meunier et al. [31] which has previously been identified as a challenging set to model using some of the currently available generalised neo-Hookean strain energy functions [9]. We will also consider two well-known additional datasets due to Kawabata et al. [32] and Jones and Treloar [33]. The tabulated data compiled and collated by us from those studies are presented in Appendix A, Tables A.1 to A.4.

3.1. Cauchy stress (σ) – stretch (λ) relationships

In case of uniaxial, equi-biaxial and pure shear deformations, the deformation gradient tensor $\mathbf{F}$ takes the forms:

$$\mathbf{F}_{uni} = \begin{bmatrix} \lambda & 0 & 0 \\ 0 & \lambda^{-1/2} & 0 \\ 0 & 0 & \lambda^{-1/2} \end{bmatrix}, \quad \mathbf{F}_{eq-bi} = \begin{bmatrix} \lambda & 0 & 0 \\ 0 & \lambda & 0 \\ 0 & 0 & \lambda^{-2} \end{bmatrix}, \quad \mathbf{F}_{ps} = \begin{bmatrix} \lambda & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & \lambda^{-1} \end{bmatrix}, \quad (17)$$

respectively. Assuming incompressibility, the second Piola-Kirchhoff stress tensor $\mathbf{S}$ is given by:

$$\mathbf{S} = 2 \frac{\partial W}{\partial \mathbf{C}} - p \mathbf{C}^{-1}, \quad (18)$$

with p as the arbitrary Lagrange multiplier. The Cauchy stress σ is derived from $\mathbf{S}$ using $\sigma = \mathbf{F} \mathbf{S} \mathbf{F}^T$, resulting in:

¹ While the original data is due to Treloar (1944), the numerical values of the nominal stress and stretch data points are those in possession of Professor Ray Ogden which were generously made available to us. From those values we compiled the Cauchy stress-stretch data for our use in this manuscript.

$$\begin{cases} \sigma_{uni} = \frac{1}{3}\mu \frac{9N - (\lambda^2 + 2\lambda^{-1})}{3N - (\lambda^2 + 2\lambda^{-1})} \left(\lambda^2 - \frac{1}{\lambda} \right), \\ \sigma_{eq-bi} = \frac{1}{3}\mu \frac{9N - (2\lambda^2 + \lambda^{-4})}{3N - (2\lambda^2 + \lambda^{-4})} \left(\lambda^2 - \frac{1}{\lambda^4} \right), \\ \sigma_{ps} = \frac{1}{3}\mu \frac{9N - (\lambda^2 + 1 + \lambda^{-2})}{3N - (\lambda^2 + 1 + \lambda^{-2})} \left(\lambda^2 - \frac{1}{\lambda^2} \right). \end{cases} \quad (19)$$

Relationships in Eq. (19) are curve-fitted onto the reported experimental data from the foregoing designated studies.

3.2. The fitting procedure

To find the set of values for μ and N that provide the best fit to all three considered deformation behaviours, we simultaneously fitted the relationships in Eq. (19) to the uniaxial, biaxial and pure shear deformation data reported in each designated study. We employed two separate approaches to minimising the residual sum of squares (RSS) function in order to find the best fits: (i) the RSS function defined as: $RSS = \sum_i (\sigma^{model} - \sigma^{experiment})_i^2$, where i is the number of data points, so as to minimise the absolute error; and (ii) the RSS function defined as: $RSS = \sum_i \left(\frac{\sigma^{model} - \sigma^{experiment}}{\sigma^{experiment}} \right)_i^2$ so as to minimise the relative error. The latter strategy was proposed to us for consideration by a reviewer. Given that the optimal values of the model parameters characterised by each strategy were found to be in broad agreement, we will report the fitting parameter values as mean $\pm$ standard deviation obtained from both methods. Both RSS minimisation methods were performed via an in-house developed code in MATLAB[®], using the genetic algorithm (GA) function. The template code and the error functions corresponding to each method have been appended as a supplementary material. Depending on the type of deformation and the number of datasets considered, the interested reader may add or adjust the $(\sigma - \lambda)$ relationships in the template code and in the error functions. The values of R^2 for each fit were calculated first by establishing the Coefficient of Correlation using a built-in MATLAB function and then squaring that value.

3.3. Fitting results

We start by presenting the results for Treloar's data. The experimental versus modelling curves are shown in Fig. 2. For the values of the model parameters see Table 1.

Fig. 2 - Experimental versus modelling plots for datasets compiled from Treloar's study [30]: (a) $\sigma - \lambda$ curves; (b) relative error (%) at each experimental data point; and (c) the Mooney plot for the datasets versus the modelling results.

From a qualitative perspective and R^2 values, plots in Fig. 2a show an excellent agreement between the model and the data. For a more quantitative analysis, we have presented the relative error (%), defined as $\frac{|\sigma^{model} - \sigma^{experiment}|}{\sigma^{experiment}} \times 100$, for each data point in Fig. 2b. While for the higher levels of deformation the relative error is low, in the lower region of deformation this trend is reversed. However, it may be worthwhile to point out that at the maximum level of relative error, i.e. for the first point in the plot related to the pure shear deformation, the absolute difference between the model prediction and the experimental data is only in the order of 0.03 MPa. In the experience of the authors, this level of variation may well fall within the expected experimental errors particularly at the initial (lower) range of deformation, compounded by defining a consistent initial undeformed position, load reading errors and other usual experimental artefacts.

Mooney plots of the datasets versus the modelling outcomes are also presented in Fig. 2c. These plots provide an additional means to compare the capability of our proposed model versus those considered in [6], which we will come back to in §4.1. Plots in Mooney space are constructed via dividing the engineering stress P by $2(\lambda - \lambda^{-2})$ presented as the y axis, and plotting the result versus λ^{-1} in the x axis. These plots are particularly useful for better illustrating the fitting results at lower stretch levels compared with the usual $\sigma - \lambda$ plots. It may be observed that the higher levels of the relative error in lower deformation regions in plots of Fig. 2b are also reflected in the Mooney plots of Fig. 2c, where the model is seen to deviate from the experimental data at higher λ^{-1} values.

Note that here we have used the three uniaxial, equi-biaxial and pure shear datasets simultaneously to find the best fit by minimising each of the considered RSS objective functions. However, the RSS function may not necessarily result in a minimum if each dataset is used individually for fitting, but instead may produce a valley. See Appendix B for more discussion on this point.

Table 1 - Fitting parameters for the considered datasets.

Treloar (1944) data			
Deformation mode	μ [MPa]	N [-]	R^2
Uniaxial	0.29 ± 0.03	26.8 ± 3.2	0.99
Biaxial			0.99
Pure shear			0.99
Kawabata et al. (1981) data			
Deformation mode	μ [MPa]	N [-]	R^2
Uniaxial	0.32 ± 0.04	36 ± 8.31	0.98
Biaxial			0.99
Pure shear			0.98
Jones and Treloar (1975) data			
Deformation mode	μ [MPa]	N [-]	R^2
Uniaxial	0.57 ± 0.15	8.5 ± 2.5	0.98
Biaxial			0.99
Pure shear			0.98

The second set of results is due to application of the model to the data from Kawabata et al. [32]. Plots in Fig. 3 compare the experimental data versus the model. Values of the model parameters are given Table 1. Compared with the fitting results in Fig. 2, we note that the relative errors in the lower stretch region of the deformation are typically smaller than those for the Treloar data. Again, we highlight that at the maximum level of relative error (pertaining to the pure shear data), the absolute difference between the model prediction and the experimental data is in the order of 0.08 MPa.

Fig. 3 - Experimental versus modelling plots for datasets collated from Kawabata et al. [32]: (a) $\sigma - \lambda$ curves; and (b) relative error (%) at each experimental data point. The inset illustrates the fits at lower stretch levels, designated by a circle in the $\sigma - \lambda$ plot.

Finally, fitting results for datasets reported by Jones and Treloar [33] are presented in Fig. 4, with the model parameters summarised in Table 1.

Fig. 4 - Experimental versus modelling plots for datasets collated from Jones and Treloar [33]: (a) $\sigma - \lambda$ curves; and (b) relative error (%) at each experimental data point. The inset illustrates the fits at lower stretch levels designated on the $\sigma - \lambda$ plot.

Note that the biaxial tests performed by Jones and Treloar are not equi-biaxial. In that particular test, rubber samples were stretched to a pre-set level of $\lambda_2 = 2.29$, and then extension was applied in λ_1 direction. Therefore, the sample will have experienced compression in λ_1 direction, prior to the application of extension. This is indeed also reflected in the starting point of λ_1 , where instead of $\lambda_1 = 1$, the deformation begins at $\lambda_1 = 0.66$. We specifically chose this dataset to test the model's capability in capturing the sample behaviour under such compression condition (Fig. 4). We also highlight that at the maximum relative error for this dataset, the absolute difference between the model prediction and the experimental data point is only in the order of 0.05 MPa.

As a means of demonstrating the capability of our proposed model versus the existing models of the same class, we now provide a brief comparison between the fits provided by our model and that of the Gent for the foregoing datasets. Both models belong to the same category of *limiting chain extensibility* generalised neo-Hookean functions, both entail a logarithmic term, and as will be shown in §4.2, both have a similar functional form. Therefore the Gent model provides a meaningful platform for comparing the fitting results with our model. Plots in Fig. 5 and Fig. 6 provide a case-by-case comparison between the two models for uniaxial, equi-biaxial and pure shear deformation data of Treloar [30] and Kawabata et al. [32], respectively. Note that the Gent model was fitted to the data using the same fitting procedure outlined in §3.2, resulting in model parameter values in the range of $\mu = 0.28 \pm 0.04$ MPa and $J_m = 86.69 \pm 15.63$ for the Treloar (1944) data, and $\mu = 0.32 \pm 0.03$ MPa and $J_m = 95.74 \pm 3.47$ for the Kawabata et al. (1981) data. From these plots it may be observed that while for the uniaxial data both models appear to produce a similar quality of fit, the Gent model produces a higher degree of error in capturing the (equi-)biaxial and pure shear deformation data. We found a similar trend for the Jones and Treloar dataset, with the relative errors falling between those reported for the Treloar [30] and Kawabata et al. [32] data in Figs. 5 and 6, and we therefore refrain from replicating the results here. An interesting outcome is also observed using the dataset of Meunier et al. [31]; *vide infra*.

Treloar (1944) data

Figure 5 – Comparison between our model and the Gent model for Treloar (1944) data: (a) uniaxial; (b) equi-biaxial; and (c) pure shear deformations. Continuous (red) lines represent the prediction of our proposed model, while the dashed (green) lines illustrate the fits provided by the Gent model. Note that the x and y axes are not to the same scale for the purpose of clarity in presentation.

Kawabata et al. (1981) data

(a)

(b)

(c)

Figure 6 – Comparison between our model and the Gent model for Kawabata et al. (1981) data: (a) uniaxial; (b) equi-biaxial; and (c) pure shear deformations. Continuous (red) lines represent the prediction of our proposed model, while the dashed (green) lines illustrate the fits provided by the Gent model. Note that the x and y axes are not to the same scale for the purpose of clarity in presentation.

It was noted in [9] that a dataset from a silicon rubber sample originally tested by Meunier et al. [31] may not be best captured by the Gent model, specifically at higher deformation ranges. We herein present the fitting results of our model to this dataset in comparison with that provided by the Gent model. The collated data points are tabulated and presented in Appendix A (Table A.4). Fig. 7 provides the fitting results and the comparison between the two models. The best fits provided by the Gent model have been illustrated by dashed green lines. Plots in Fig. 7d represent the relative error at each data point for both models. It may be observed that the Gent model encounters difficulties capturing the whole data, particularly for the larger deformation regimes in equi-biaxial and pure shear datasets, compared with our proposed model. Note the similarity in this trend with the Treloar and Kawabata et al. datasets presented in Figs. 5 and 6. The values for the model parameters are summarised in Table 2.

Table 2 - Fitting parameters for the dataset due to Meunier et al. [31].

Meunier et al. (2008) data			
Deformation mode	μ [MPa]	N [-]	R^2
Uniaxial			0.99
Biaxial	0.27 ± 0.01	4.49 ± 0.60	0.99
Pure shear			0.99

Fig. 7 - Experimental versus modelling plots for datasets due to Meunier et al. [31]: (a) uniaxial; (b) biaxial; (c) pure shear deformations; and (d) relative errors. Continuous (red) lines represent the prediction of our proposed model, while the dashed (green) lines illustrate the fits provided by the Gent model with $\mu = 0.333 \pm 0.004$ MPa and $J_m = 15.62 \pm 0.34$. Note that the scale of the y axis in panel (c) is not the same as that in panels (a) and (b) for clarity in presentation.

4. Discussion and analysis

Rubber elasticity literature has a rich history of well-established models successfully applied to the finite deformation of rubbers and elastomers. In the realm of generalised neo-Hookean functions, models presented by Arruda and Boyce [22] and Gent [13], to name but a few, have provided valuable contributions and major advancements in this field. The success of these models, however, does not impede the motivation based on which we have developed and put forward our proposed model herein: (i) its simplicity in that the model contains only two parameters, namely μ and N ; (ii) its direct structural roots in molecular/statistical mechanics principles; and (iii) providing modelling consistency, at least for pure homogenous deformations, where the model seems to be able to fit well to the considered deformation modes with a single set (or a very narrow range of variation) of parameters. We also note the improved quality of fits provided by our proposed model to the considered datasets compared with that of the Gent, as demonstrated in §3.3.

The model presented here was previously put forward by us in a different context, for use in modelling the deformation of the isotropic matrix of the aortic heart valve tissue [3]. Here, we have extended the application of the model to the finite deformation of rubbers and elastomers. While preparing this manuscript, an article by the late Michael Carroll was published posthumously, who independently from us had developed a similar model in 2010², but only published [34] more than a year after our original model in [3]. The model presented in [34], equation (7.5) therein, is similar in form to our model, with a slight variation arising from a different method of approximation. However, the background, rationale and approach used in this study are independent and distinct from that of [34].

² Private communication with Professor Michel Destrade.

4.1. Quality of fits provided by the proposed model

Let us first start by noting the results in Table 1. A narrow range for μ and N , rather than a single value, has been presented in that table and throughout this paper. In view of considering two different RSS functions as outlined in §3.2, which their respective minimisation leads to slightly different values of μ and N , as well as potential variations in data collation and the experimental errors in the original studies, reporting this (narrow) range was deemed more reasonable and comprehensive as opposed to providing a single value. For Treloar [30] and Kawabata et al. [32] datasets, the variation range is an order of magnitude lower than the output value of the fit. Therefore, the reported values may well be considered as effectively a single value for each dataset. However, we note that the dataset due to Jones and Treloar [33] appears to be the more challenging set to model, as μ for this dataset possesses a wider range than the other two datasets, and the variation of N is in the same order of magnitude as the provided average. From a continuum mechanics point of view, however, there may not be a significant difference between the number of Kuhn segments $N = 8.5$ and $N = 11$. Kuhn segment is not a clear-cut geometrical property, and a level of arbitration may be involved in segmentation. The variation in values of μ results from those differences in N values. Therefore, if we reasonably accept that $N = 8.5$ is notionally close to $N = 11$, the variation range of ± 0.15 units in μ values may not be deemed significant in this context. Therefore, within the considered deformation modes for each specimen, the model appears to provide an excellent consistency with just a single set of parameter values (with a narrow range of variation). In order to illustrate the robustness of the results and the stability of the fits provided by our proposed model, a concise sensitivity analysis of the model fittings to perturbed Treloar [30], Kawabata et al. [32] and Jones and Treloar [33] datasets, to the effect of $\pm 10\%$ of the (Cauchy) stress, has been presented in Appendix C. The fitting results indicate that such a relatively high level of perturbation does not inflict an instability in the fitting results, where in particular the values of N remain stable.

Let us now consider the Mooney plots in Fig. 2c. Following [6] we have provided these plots for Treloar data [30] to offer another means for a direct comparison between our model and the neo-Hookean and the Yeoh models considered therein. The assessment of the Mooney plots in [6] and herein readily reveals the better quality of fits provided by our model. However, we also note that for lower ranges of deformation, the Mooney plots in Fig. 2c indicate deviations between the model and the experimental data points, as also reflected in plots of the relative

error in Fig. 2b. Both plots, however, confirm favourable fittings at higher levels of deformation.

On this note, the addition of I_2 terms to the model, apart from addressing the requirement of compatibility with fourth-order weak nonlinear elasticity and satisfying universal relationships, will likely also improve the fitting quality. Should such a modification be required, the literature indicates a number of adjustments incorporated into the existing generalised neo-Hookean models, such as the one proposed by Pucci and Saccomandi [35], suggesting the addition a logarithmic function of I_2 to the original Gent model. We further note that within the range of deformation considered in some of the experiments, particularly at the higher levels of stretch/deformation, it is possible for molecular linkages and bonds to irreversibly break, resulting in residual stretches and damage, and/or other inelastic behaviours (see, e.g., [36]). Generalised neo-Hookean functions, including our proposed model, do not fundamentally capture or allow for this feature, which otherwise may too improve the quality of fits.

For the data presented in §3.3, we note the plots in Fig. 5 and Fig. 6 for the uniaxial, (equi-)biaxial and pure deformation datasets due to Treloar [30] and Kawabata et al. [32], respectively. Those plots demonstrate the improved fits provided by our proposed model compared with those of the Gent model, particularly for the (equi-)biaxial and pure shear deformations (panels (b) and (c) in those plots). The plots in Fig. 5c and Fig. 6c typically show lower relative errors produced by the proposed model versus that of the Gent. A similar trend was also observed for Jones and Treloar [33] data, and as illustrated in Fig. 7, was repeated again using the Meunier et al. [31] dataset particularly at higher ranges of deformation. As a hypothesis at this point, the reason why our proposed model provides a better description of the biaxial and pure shear data, and at higher deformations, may lie in the way that the inverse Langevin function has been approximated. On using the analysis provided by Horgan and Saccomandi [26], we note that our proposed model is a higher order constrained Padé approximation of the inverse Langevin function. The Gent model is a rational Padé approximant of order $[0/1]$, while our model is of $[1/1]$ order [26]. Given that the strain-stiffening effect is a direct consequence of the inverse Langevin function behaviour, a more accurate approximation of this function results in a better capturing of the deformation behaviour of the samples in stiffer regimes/regions. We also note from the values of model parameters in Table 2 that our proposed model yet again provides an excellent mechanistic

modelling consistency in describing the deformation behaviour of the Meunier et al. [31] dataset, as all deformation modes are captured with a single set of μ and N values (with a very narrow range of values).

4.2. A brief comparison with other generalised neo-Hookean models

It may prove insightful to provide a qualitative comparison between our proposed model herein and the landmark generalised neo-Hookean models of Arruda-Boyce and of Gent, as well as a brief mathematical comparison between the functional form of those models and ours, and that of the Lopez-Pamies [9].

4.2.1. Qualitative comparisons

The seminal work of Arruda and Boyce [22] presented a model based on a non-Gaussian statistical mechanics treatment of the freely jointed molecular chains. In their model, the ensuing inverse Langevin function was approximated by a (truncated) Taylor series expansion, previously presented by Treloar [37]. As also described elsewhere (e.g., see [5]), the Taylor series expansion approximation eliminates the asymptotic limiting chain extension characteristic of the inverse Langevin function. This characteristic would be an additional structural feature which will be accounted for by using our proposed model, while it would be lost in the Arruda-Boyce model. However, and perhaps more pertinent than this feature, is another drawback as a result of Taylor series expansion approximation. The original inverse Langevin function $\ell^{-1}(x)$ is only valid within the domain $-1 < x < 1$. This imposes the *a priori* requirement $N > I_1/3$ (see §2.2). When the Taylor series expansion is used, however, this *a priori* requirement vanishes since the inverse Langevin function is replaced by a polynomial. There exist experimental datasets for which the Arruda-Boyce model only provides the best fit with numerical values of N that do not satisfy the *a priori* requirement, and therefore are not mathematically valid. A detailed example and analysis on this point is presented in [3]. No such issue arises when using our proposed model.

Another landmark generalised neo-Hookean model is that of Gent [13], originally proposed as a phenomenological constitutive relationship for rubberlike materials. The model has a simple form and has proved proficient in fitting to various experimental datasets. The literature

highlights efforts to provide a mathematical underpinning for the molecular basis of the model (see, e.g., Puglisi and Saccomandi [11] and Horgan [12]); however the Gent model lacks a direct structural basis and derivation. In addition, comparing the fits provided by the Gent model and our proposed model to the dataset considered in §3.3, it may readily be observed that our model provides improved fits compared with the Gent model, particularly for biaxial and pure shear datasets. Therefore, while the authors are not positioned to provide the fitting results for a wider variety of different types/classes of elastomers, based on the considered datasets our proposed model may be deemed a well-suited choice.

Considering the models beyond the generalised neo-Hookean classification, a recurring feature may be observed as the presence of material constants without a direct physical interpretation whose values can only be known *a posteriori* after data fitting, or alternatively with the presence of a relatively high number of model constants. An objective evaluation of such models in relation to the issues surrounding the uniqueness of the fits and optimal solutions is provided by Ogden et al. [7], where two distinct points have been identified: (i) it is possible to obtain more than one optimal set of model parameters when using such models to fit the experimental data; and (ii) the optimal values of the model parameters, and the fitting process as a whole, becomes very sensitive to the numerical method being employed. Both these points give rise to issues of uniqueness and the objectivity of the quantified parameters. Our proposed model in comparison is simple as it embodies only two model parameters which have physical definitions and objectivity. It may be humorous to underpin this point by reciting a famous quote attributed to John von Neumann: “With four parameters I can fit an elephant, and with five I can make him wiggle his trunk”³. One should imagine that with only two parameters there won’t be too much room for von Neumann’s elephant for manoeuvre.

4.2.2. A mathematical comparison between the functional forms

Let us start by recalling the Arruda-Boyce strain energy function W_{AB} :

$$W_{AB} = \mu \left[\frac{1}{2} (I_1 - 3) + \frac{1}{20N} (I_1^2 - 9) + \frac{11}{1050N^2} (I_1^3 - 27) + \frac{19}{7000N^3} (I_1^4 - 81) + \dots \right], \quad (20)$$

which can simply be presented in the series form as:

³ *Nature*, volume 427, page 297 (22 January 2004)

$$W_{AB} = \sum_{i=1}^n C_i (I_1^i - 3^i) . \quad (21)$$

It is straightforward to show that the Taylor series expansion of the Gent model W_G yields:

$$W_G = \frac{\mu}{2} \left[(I_1 - 3) + \frac{1}{2J_m} (I_1 - 3)^2 + \frac{1}{3J_m^2} (I_1 - 3)^3 + \frac{1}{4J_m^3} (I_1 - 3)^4 + \dots \right] , \quad (22)$$

which is equivalent to:

$$W_G = \sum_{i=1}^n C_i (I_1 - 3)^i . \quad (23)$$

Finally, the model presented by Lopez-Pamies [9] takes the general W_{L-P} form:

$$W_{L-P} = \sum_{i=1}^n C_i (\alpha_i) (I_1^{\alpha_i} - 3^{\alpha_i}) , \quad (24)$$

where α_i s are real-valued parameters.

Now, the Taylor expansion of our proposed model may be written as:

$$W = \mu N \left[\frac{1}{6N} (I_1 - 3) + \frac{I_1 - 3}{3N - 3} + \frac{(I_1 - 3)^2}{2(3N - 3)^2} + \frac{(I_1 - 3)^3}{3(3N - 3)^3} + \dots \right] = \sum_{i=1}^n C_i (I_1 - 3)^i . \quad (25)$$

These series representations of the functional forms of the considered strain energy functions provide a useful unified tool to directly compare the mathematics of the proposed models. In this regard, comparing Eqs. (21) and (23-25) makes some interesting observations. First, it is clear that the functional form of our model is similar to that of the Gent but distinct from the Arruda-Boyce and Lopez-Pamies models, whereas the Arruda-Boyce and Lopez-Pamies models have a similar functional form. Therefore, while the starting point for deriving our model is similar to that of the Arruda and Boyce model, the final functional forms of the two models are different. Second, while Eqs. (21) and (24) indicate a similarity between the functional forms of Arruda-Boyce and Lopez-Pamies models, we note that the power terms in the former are positive integers while in the latter they are real-valued. For all the structurally-relevant models considered herein, i.e. the Arruda-Boyce, Gent and our proposed models, the power terms are positive integers. However, the model of Lopez-Pamies [9] was proposed as a phenomenological model. A reviewer also brought to our attention a generalised version of the Yeoh model in the form of $\sum_{i=1}^3 C_i (I_1 - 3)^{\alpha_i}$, presented in [38], where again α_i terms are real valued. Whereas the C_i constants in the Arruda-Boyce, Gent and our proposed models have

their roots in structural parameters, the arbitrary nature of the values that α_i can assume in the phenomenological Lopez-Pamies and generalised Yeoh models makes it difficult to envisage a direct structural meaning for this parameter. However, from a curve-fitting point of view, the presence of the α_i parameters in those models renders them versatile functions to better capture intricate experimental datasets such as that of Lahellec et al. [39] by the Lopez-Pamies model [9] or the datasets considered in [38] using the generalised Yeoh model.

4.3. A note regarding I_2 terms

As highlighted in [19], the inclusion of an I_2 term in a strain energy function W in general provides a better modelling of the experimental data. However, more importantly, instructive studies such as [17], later revisited and elaborated further in [5,6,18], demonstrate that generalised neo-Hookean functions, independent of their functional form, are not adequate to capture the full deformation modes of elastomers. Specific examples have been provided in [17] and [18] in reference to torsional deformation, and for simple shear in [19], where it has been shown that the dependence of the W function on I_2 is necessary. In particular, a useful experimental protocol was devised in [40] to check the validity of considering only I_1 terms in the strain energy function W . By drilling a hole in one of the plates attached to a specimen under simple shear deformation test, it is possible to observe whether the surface of the specimen will tend to bulge or remain flat. The assumption of $W = f(I_1)$ implies that $\sigma_{22} = 0$ in simple shear, and therefore bulging of the specimen surface will be indicative of $\sigma_{22} \neq 0$ so that an I_2 term is required. In addition, from the requirements of compatibility with fourth-order weak nonlinear elasticity, the inclusion of a C_2 term in the strain energy function W may also be essential. Therefore, a valid and universal model must include C_2 and I_2 terms in its functional form.

Generalised neo-Hookean functions, however, by definition only depend on I_1 . These functions, while not providing universality, offer mathematical simplicity and provide sufficiently good quality of fits for a range of elastomers and deformation modes. For such elastomers and within the modes of deformation that a generalised neo-Hookean function may address, a body of literature in rubber elasticity suggests that $\frac{\partial W}{\partial I_1}$ is generally much greater than $\frac{\partial W}{\partial I_2}$, and therefore neglecting $\frac{\partial W}{\partial I_2}$ may not introduce serious fitting errors [41]. It has been

highlighted that I_2 in general has a significant correlation with I_1 , and thus when fitting a generalised neo-Hookean function to the experimental data, $\frac{\partial W}{\partial I_2}$ is not completely neglected, but rather part of that information is included in $\frac{\partial W}{\partial I_1}$ [41]. A reviewer highlighted other studies including those by Davies et al. [42] and the references therein, which experimentally verify the assumption of neglecting the I_2 term in modelling the planar deformation of rubber samples up to strains of 100%. In addition, the origins of the C_2 term in rubber elasticity are thought to be linked with the non-affine deformation of the entanglement network formed by physical and chemical crosslinks [20]. Providing a theoretical and structural underpinning for C_2 terms may therefore undermine the mathematical simplicity offered by the generalised neo-Hookean strain energy functions. For these reasons, we note that generalised neo-Hookean functions, including our proposed model, are a subset and a convenient simplification of what may be considered as a universally valid model.

5. Concluding remarks

A new model within the class of generalised neo-Hookean strain energy functions was presented. With only two model parameters, namely μ and N , it was shown that our proposed model provides improved fits to the considered experimental data compared with the existing models of the same class in the literature, particularly for biaxial and pure shear deformations. Other advantages include direct structural and molecular origins of the model, simplicity of the fitting procedure, and ability of the model to provide a good fit to many deformation modes of a specimen with a single set of parameters (or within a very narrow range). Where applicable, terms containing C_2 and I_2 may also be included in the model to further improve its predictions. Given these advantages, we propose this model for application to the deformation of rubber-like and elastomeric materials. While not explored in this manuscript, we draw the attention of the reader to another potential ground for application of our proposed model, where it may serve to represent the contribution of the isotropic matrix in modelling the large deformation of soft biological tissues.

Acknowledgment

The authors are grateful to Professor Giuseppe Saccomandi for valuable discussions and suggestions to improve this work. We also extend our gratitude to the reviewers for their constructive remarks which have been implemented in the final version of this manuscript.

Appendix A: Tabulated experimental data

Table A.1 - Experimental data compiled from Treloar (1944) [30].

Uniaxial		Biaxial		Pure Shear	
λ [-]	σ [MPa]	λ [-]	σ [MPa]	λ [-]	σ [MPa]
1.00	0	1	0	1	0
1.02	0.03	1.03	0.095	1.03	0.07
1.125	0.15	1.065	0.17	1.12	0.18
1.24	0.28	1.115	0.265	1.19	0.29
1.39	0.44	1.14	0.29	1.31	0.435
1.585	0.65	1.20	0.39	1.46	0.61
1.90	0.95	1.31	0.57	1.84	1.09
2.18	1.28	1.42	0.72	2.38	1.82
2.42	1.64	1.68	1.09	2.97	2.78
3.02	2.61	1.94	1.48	3.47	3.88
3.57	3.75	2.49	2.39	3.95	5.08
4.03	4.94	3.03	3.755	4.37	6.39
4.76	7.56	3.43	4.94	4.69	7.64
5.36	10.46	3.75	6.40	4.97	8.97
5.75	13.31	4.07	8.03		
6.15	16.53	4.26	9.38		
6.40	19.46	4.45	10.76		
6.60	22.53				
6.85	25.87				
7.05	29.12				
7.15	32.12				
7.25	35.28				
7.40	38.69				
7.50	41.94				
7.60	48.01				

Table A.2 - Experimental data due to Kawabata et al. (1981) [32].

Uniaxial		Biaxial		Pure Shear	
λ [-]	σ [MPa]	λ [-]	σ [MPa]	λ [-]	σ [MPa]
1.00	0	1.00	0	1.00	0
1.04	0.045	1.04	0.088	1.04	0.061
1.06	0.065	1.06	0.129	1.06	0.091
1.08	0.093	1.08	0.178	1.08	0.122
1.10	0.117	1.10	0.218	1.10	0.152
1.12	0.140	1.12	0.257	1.12	0.181
1.14	0.163	1.14	0.293	1.14	0.209
1.16	0.186	1.16	0.329	1.16	0.238
1.20	0.235	1.20	0.396	1.20	0.294
1.24	0.282	1.24	0.461	1.24	0.349
1.30	0.351	1.30	0.547	1.30	0.434
1.60	0.693	1.60	0.976	1.60	0.819
1.90	1.051	1.90	1.414	1.90	1.193
2.20	1.432	2.20	1.896	2.20	1.593
2.50	1.850	2.50	2.435	2.50	2.022
2.80	2.316	2.80	3.030	2.80	2.482
3.10	2.849	3.10	3.689	3.10	2.973
3.40	3.454			3.40	3.505
3.70	4.133			3.70	4.077

Table A.3 - Experimental data collated from Jones and Treloar (1975) [33].

Uniaxial		Biaxial		Pure Shear	
λ [-]	σ [MPa]	λ [-]	σ [MPa]	λ [-]	σ [MPa]
1.00	0	0.66	0.00	1.00	0.00
1.02	0.04	0.71	0.05	1.04	0.08
1.04	0.07	0.78	0.15	1.08	0.16
1.08	0.14	0.87	0.30	1.15	0.32
1.13	0.20	1.00	0.52	1.21	0.44
1.17	0.25	1.19	0.93	1.30	0.65
1.31	0.44	1.35	1.35	1.46	1.10
1.53	0.76	1.53	1.92	1.71	1.90
1.71	1.00	1.72	2.68	1.96	2.96
1.96	1.38	1.96	3.84	2.27	4.67
2.21	1.75	2.12	4.72	2.52	6.35
2.35	2.03	2.28	5.83	2.80	8.54
2.47	2.28				
2.62	2.55				
2.79	2.95				
2.92	3.29				

Table A.4 - Experimental data due to Meunier et al. (2008) [31].

Uniaxial		Biaxial		Pure Shear	
λ [-]	σ [MPa]	λ [-]	σ [MPa]	λ [-]	σ [MPa]
0.49	-0.68	1	0	1	0
0.52	-0.62	1.05	0.10	1.02	0.05
0.55	-0.56	1.10	0.20	1.05	0.10
0.58	-0.51	1.18	0.32	1.10	0.17
0.61	-0.46	1.23	0.41	1.15	0.23
0.64	-0.41	1.34	0.54	1.22	0.30
0.67	-0.37	1.44	0.69	1.29	0.39
0.71	-0.33	1.54	0.83	1.38	0.48
0.74	-0.29	1.68	1.09	1.48	0.59
0.77	-0.26	1.77	1.31	1.57	0.71
0.80	-0.22	1.80	1.39	1.66	0.83
0.83	-0.18	1.84	1.52	1.75	0.96
0.86	-0.15	1.95	1.97	1.83	1.10
0.89	-0.11	2.06	2.72	1.90	1.24
0.93	-0.08			1.97	1.38
0.96	-0.04			2.02	1.51
1.00	0			2.07	1.66
1.05	0.05			2.12	1.80
1.11	0.11			2.15	1.94
1.18	0.17				
1.27	0.25				
1.36	0.33				
1.46	0.44				
1.56	0.54				
1.66	0.66				
1.76	0.78				
1.85	0.91				
1.93	1.06				
1.99	1.18				
2.04	1.32				
2.09	1.45				
2.14	1.60				
2.17	1.73				

Appendix B: The residual sum of squares (RSS) function in simultaneous versus individual fittings

As discussed earlier, we have found the optimal set of μ and N by simultaneously fitting our model to the three uniaxial, biaxial and pure shear datasets, where either of the considered RSS functions in this study, i.e. $RSS = \sum_i (\sigma^{model} - \sigma^{experiment})_i^2$ or $RSS = \sum_i \left(\frac{\sigma^{model} - \sigma^{experiment}}{\sigma^{experiment}} \right)_i^2$, were minimised by considering all the datasets. By means of illustration, let us consider the former definition of the RSS function. The plot of this RSS function as a result of our fitting to the Treloar data is shown in Figure B.1. It may be observed that the function has a clear minimum, obtained at $\mu = 0.28$ MPa and $N = 26.44$.

Fig. B.1 - The residual sum of squares (RSS) function defined as $\sum_i (\sigma^{model} - \sigma^{experiment})_i^2$ plotted versus the model parameters μ and N for Treloar [30] data, when all three uniaxial, biaxial and pure shear data are used simultaneously to obtain the best fit.

However, if only any one of the datasets is used, the RSS function may not necessarily exhibit minima, but rather it may result in a valley. We undertook to plot the RSS function by finding the best fit using separately the uniaxial, biaxial and pure shear data of Treloar [30]. The results

are plotted in Fig. B.2. It may be observed that while fitting our model to only uniaxial or biaxial data may result in the best fit through a full minimum, if the pure shear data were used no unique set of optimal μ and N could be found as the RSS function exhibits a clear valley along N . The same observation also holds true for the other definition the RSS function. We conclude that for a more robust optimisation and a higher degree of confidence in finding the optimal set of model parameters, it may prove desirable to fit the model simultaneously to various sets of deformation data, as has been undertaken in this study.

Fig. B.2 - The residual sum of squares (RSS) function defined as $\sum_i (\sigma^{model} - \sigma^{experiment})_i^2$ plotted versus the model parameters μ and N using Treloar [30] data for: (a) uniaxial; (b) biaxial; and (c) pure shear data. Note that each dataset has been considered separately for each fit. The RSS function clearly

possesses minima in the case of uniaxial and biaxial data fittings; however note the valley in the case of pure shear.

Appendix C: Modelling results for perturbed datasets

In order to provide a measure of sensitivity analysis for our fitting results, here we present the fitting of our model to perturbed datasets from Treloar [30], Kawabata et al. [32] and Jones and Treloar [33], in Figs. C.1, C.2 and C.3, respectively. The perturbation has been introduced to the tune of $\pm 10\%$ of the original (Cauchy) stress data points as tabulated in Appendix A. Values of the model parameters for each fit have been illustrated on the plots. The results indicate that the robustness of the fits is intractable to such large perturbations and the characterised values of the parameters remain stable.

Figure C.1 - Perturbed data of the original Treloar [30] study. Note that the parameter values for the original data are $\mu = 0.29 \pm 0.03$ MPa and $N = 26.80 \pm 3.20$. The axes are not the same scale in all plots for the clarity of presentation.

Figure C.2 - Perturbed data of the original Kawabata et al. [32] study. Note that the parameter values for the original data are $\mu = 0.32 \pm 0.04$ MPa and $N = 36 \pm 8.31$.

Figure C.3 - Perturbed data of the original Jones and Treloar [33] study. Note that the parameter values for the original data are $\mu = 0.57 \pm 0.15$ MPa and $N = 8.50 \pm 2.50$. The y axis is not the same scale in all plots for the clarity of presentation.

References

- [1] G.A. Holzapfel, T.C. Gasser, R.W. Ogden, A new constitutive framework for arterial wall mechanics and a comparative study of material models, *J. Elasticity* 61 (2000) 1-48.
- [2] A. Anssari-Benam, A. Bucchi, H.R.C. Screen, S.L. Evans, A transverse isotropic viscoelastic constitutive model for the aortic valve tissue, *R. Soc. Open Sci.* 4 (2017) 160585, doi: 10.1098/rsos.160585.
- [3] A. Anssari-Benam, A. Bucchi, Modelling the deformation of the elastin network in the aortic valve, *J. Biomech. Eng.* 140 (2018) 011004, doi: 10.1115/1.4037916.
- [4] A. Anssari-Benam, Y.-T. Tseng, A. Bucchi, A transverse isotropic constitutive model for the aortic valve tissue incorporating rate-dependency and fibre dispersion: Application to biaxial deformation, *J. Mech. Behav. Biomed. Mater.* 85 (2018) 80-93.
- [5] G. Puglisi, G. Saccomandi, Multi-scale modelling of rubber-like materials and soft tissues: an appraisal, *Proc. R. Soc. A* 472 (2016) 20160060, doi: 10.1098/rspa.2016.0060.
- [6] M. Destrade, G. Saccomandi, I. Sgura, Methodical fitting for mathematical models of rubber-like materials, *Proc. R. Soc. A* 473 (2017) 20160811, doi: 10.1098/rspa.2016.0811.
- [7] R.W. Ogden, G. Saccomandi, I. Sgura, Fitting hyperelastic models to experimental data, *Comput. Mech.* 34 (2004) 484-502.
- [8] O.H. Yeoh, Characterisation of elastic properties of Carbon-black-filled rubber vulcanizates, *Rubber Chem. Technol.* 63 (1990) 792-805.
- [9] O. Lopez-Pamies, A new I_1 -based hyperelastic model for rubber elastic materials, *C. R. Mecanique* 338 (2010) 3-11.
- [10] O.H. Yeoh, P.D. Fleming, A new attempt to reconcile the statistical and phenomenological theories of rubber elasticity, *J. Polym. Sci. B* 35 (1997) 1919-1931.

- [11] G. Puglisi, G. Saccomandi, The Gent model for rubber-like materials: an appraisal for an ingenious and simple idea, *Int. J. Non Linear Mech.* 68 (2015) 17-24.
- [12] C.O. Horgan, The remarkable Gent constitutive model for hyperelastic materials, *Int. J. Non Linear Mech.* 68 (2015) 9-16.
- [13] A.N. Gent, A new constitutive relation for rubber, *Rubber Chem. Technol.* 69 (1996) 59-61.
- [14] A. Bucchi, G.E. Hearn, Predictions of aneurysm formation in distensible tubes: Part A - Theoretical background to alternative approaches, *Int. J. Mech. Sci.* 71 (2013) 1-20.
- [15] R.W. Ogden, On isotropic tensors and elastic moduli, *Proc. Camb. Phil. Soc.* 75 (1974) 427-436.
- [16] M. Destrade, R.W. Ogden, On the third- and fourth-order constants of incompressible isotropic elasticity. *J. Acoust. Soc. Am.* 128 (2010) 3334-3343.
- [17] C.O. Horgan, G. Saccomandi, Simple torsion of isotropic, hyperelastic, incompressible materials with limiting chain extensibility, *J. Elasticity* 56 (1999) 159-170.
- [18] A. Wineman, Some results for generalized neo-Hookean elastic materials, *Int. J. Nonlin. Mech.* 40 (2005) 271-279.
- [19] C.O. Horgan, M.G. Smayda, The importance of the second strain invariant in the constitutive modeling of elastomers and soft biomaterials, *Mech. Mater.* 51 (2012) 43-52.
- [20] M.H. Wagner, The origin of the C_2 term in rubber elasticity, *J. Rheol.* 38 (1994), 655-670.
- [21] H.M. James, E. Guth, Theory of the elastic properties of rubber, *J. Chem. Phys.* 11 (1943) 455-481.
- [22] E.M. Arruda, M.C. Boyce, A three-dimensional constitutive model for the large stretch behavior of rubber elastic materials, *J. Mech. Phys. Solids* 41 (1993) 389-412.

- [23] M.F. Beatty, An average-stretch full-network model for rubber elasticity, *J. Elasticity* 70 (2003) 65-86.
- [24] C. Miehe, S. Göktepe, F. Lulei, A micro-macro approach to rubber-like materials - Part I: the non-affine micro-sphere model of rubber elasticity, *J. Mech. Phys. Solids* 52 (2004) 2617-2660.
- [25] A. Cohen, A Padé approximant to the inverse Langevin function, *Rheol. Acta* 30 (1991) 270-273.
- [26] C.O. Horgan, G. Saccomandi, A molecular-statistical basis for the Gent constitutive model of rubber elasticity, *J. Elasticity* 68 (2002) 167-176.
- [27] C.O. Horgan, G. Saccomandi, Phenomenological hyperelastic strain-stiffening constitutive models for rubber, *Rubber Chem. Technol.* 79 (2006) 152-169.
- [28] J.M. Ball, Convexity conditions and existence theorems in nonlinear elasticity, *Arch. Ration. Mech. An.* 63 (1976) 337-403.
- [29] J. Schröder, P. Neff, Invariant formulation of hyperelastic transverse isotropy, *Int. J. Solids Struct.* 40 (2003), 401-445.
- [30] L.R.G. Treloar, Stress-strain data for vulcanised rubber under various types of deformation, *Trans. Faraday Soc.* 40 (1944) 59-70.
- [31] L. Meunier, G. Chagnon, D. Favier, L. Orgeas, P. Vacher, Mechanical experimental characterisation and numerical modelling of an unfilled silicone rubber, *Polym. Test.* 27 (2008) 765-777.
- [32] S. Kawabata, M. Matsuda, K. Tei, H. Kawai, Experimental survey of the strain energy density function of isoprene rubber vulcanizate, *Macromolecules* 14 (1981) 154-162.
- [33] D.F. Jones, L.R.G. Treloar, The properties of rubber in pure homogeneous strain, *J. Phys. D: Appl. Phys.* 8 (1975) 1285-1304.

- [34] M.M. Carroll, Molecular chain networks and strain energy functions in rubber elasticity, *Phil. Trans. R. Soc. A* 377 (2019) 20180067, doi: 10.1098/rsta.2018.0067.
- [35] E. Pucci, G. Saccomandi, A note on the Gent model for rubber-like materials, *Rubber Chem. Technol.* 75 (2002) 839-852.
- [36] D. De Tommasi, G. Puglisi, E. Toma, F. Trentadue, A predictive model for the hysteretic and damage behavior of rubberlike materials, *J. Rheol.* 63 (2019) 1-10.
- [37] L.R.G. Treloar, The photoelastic properties of short-chain molecular networks, *Trans. Faraday Soc.* 50 (1954) 881-896.
- [38] T. W. Hohenberger, R.J. Windslow, N.M. Pugno, J.J.C. Busfield, A constitutive model for both low and high strain nonlinearities in highly filled elastomers and implementation with user-defined material subroutines in ABAQUS, *Rubber Chem. Technol.* 92 (2019) 653-686.
- [39] N. Lahellec, F. Mazerolle, J.C. Michel, Second-order estimate of the macroscopic behaviour of periodic hyperelastic composites: Theory and experimental validation, *J. Mech. Phys. Solids* 52 (2004) 27-49.
- [40] M. Destrade, M.D. Gilchrist, J.G. Murphy, B. Rashid, G. Saccomandi, Extreme softness of brain matter in simple shear, *Int. J. Non Linear Mech.* 75 (2015) 54-58.
- [41] O.H. Yeoh, Some forms of the strain energy function for rubber, *Rubber Chem. Technol.* 66 (1993) 754-771.
- [42] C.K.L. Davies, D.K. De, A.G. Thomas, Characterization of the behavior of rubber for engineering design purposes. 1. stress-strain relations, *Rubber Chem. Technol.* 67 (1994), 716-728.