

Generalised invariants and pseudo-universal relationships for hyperelastic materials: A new approach to constitutive modelling

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Abstract

Constitutive modelling of nonlinear isotropic elastic materials requires a general formulation of the strain-energy function in terms of invariants, or equivalently in terms of the principal stretches $\{\lambda_1, \lambda_2, \lambda_3\}$. Yet, when choosing a particular form of a model, the representation in terms of either the principal invariants or stretches becomes important, since a judicious choice between one or the other can lead to a better encapsulation and interpretation of much of the behaviour of a given material. Here, we introduce a family of generalised isotropic invariants, including a member $\mathcal{J}_\alpha = \lambda_1^\alpha + \lambda_2^\alpha + \lambda_3^\alpha$, which collapses to the classical first and second invariant of incompressible elasticity when α is 2 or -2, respectively. Then, we consider incompressible materials for which the strain-energy can be approximated by a function W that solely depends on this invariant \mathcal{J}_α . A natural question is to find α that best captures the finite deformation of a given material. We first show that there exist pseudo-universal relationships that are independent of the choice of W , and which only depend on α . Then, on using these pseudo-universal relationships, we show that one can obtain the exponent α that best fits a given dataset before seeking a functional form for the strain-energy function W . This two-step process delivers the best model that is a function of a single invariant. We show, on using specific examples, that this procedure leads to an excellent and easy to use approximation of constitutive models.

Keywords:

Hyperelasticity, constitutive modelling, generalised isotropic invariant \mathcal{J} , pseudo-universal relationships.

1. Introduction

In isotropic hyperelasticity, the central concept in describing the deformation is the deformation gradient \mathbf{F} . From this deformation gradient, we define the left $\mathbf{B} = \mathbf{F}\mathbf{F}^T$ and right $\mathbf{C} = \mathbf{F}^T\mathbf{F}$ Cauchy-Green deformation tensors. Due to objectivity and isotropy, we know that the strain-energy function W can then be written as a function of either the principal stretches $\{\lambda_1, \lambda_2, \lambda_3\}$; i.e., the square roots of the eigenvalues of \mathbf{B} as in the work of Valanis and Landel (1967), or any other symmetric combination of these stretches such as the classical invariants of \mathbf{B} used, for example, in the seminal work of Rivlin (1948). Clearly, any other set of isotropic invariants based on λ_i are also admissible as, for example, within the framework of weakly-nonlinear theory of elasticity where the preferred choice of strain measure is the Green – Lagrangian strain tensor $\mathbf{E} = (\mathbf{C} - \mathbf{I})/2$, and thus the set of invariants $\{\text{trace } \mathbf{E}, \text{trace } \mathbf{E}^2, \text{trace } \mathbf{E}^3\}$ is used (see, e.g.,

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Saccomandi and Vergori, 2021).

Mathematically, any choice of *complete* set of invariants is equivalent. For example, for isotropic materials, the strain-energy function can be expressed as a function of the principal stretches $W_{\text{ps}} = W_{\text{ps}}(\lambda_1, \lambda_2, \lambda_3)$, where the subscript ‘ps’ denotes principal stretches, with the symmetry condition:

$$W_{\text{ps}}(\lambda_1, \lambda_2, \lambda_3) \equiv W_{\text{ps}}(\lambda_2, \lambda_3, \lambda_1) \equiv W_{\text{ps}}(\lambda_3, \lambda_1, \lambda_2) . \quad (1)$$

Another popular choice is to use the complete set of (principal) invariants of \mathbf{B} :

$$I_1 = \text{tr}(\mathbf{B}) = \lambda_1^2 + \lambda_2^2 + \lambda_3^2, \quad (2)$$

$$I_2 = \frac{1}{2} [(\text{tr}(\mathbf{B}))^2 - \text{tr}(\mathbf{B}^2)] = \lambda_1^2 \lambda_2^2 + \lambda_2^2 \lambda_3^2 + \lambda_1^2 \lambda_3^2, \quad (3)$$

$$I_3 = \det(\mathbf{B}) = \lambda_1^2 \lambda_2^2 \lambda_3^2. \quad (4)$$

Alternatively, one may use the not-so-popular set:

$$i_1 = \lambda_1 + \lambda_2 + \lambda_3, \quad (5)$$

$$i_2 = \lambda_1 \lambda_2 + \lambda_2 \lambda_3 + \lambda_1 \lambda_3, \quad (6)$$

$$i_3 = \lambda_1 \lambda_2 \lambda_3, \quad (7)$$

or more generally any other set of three invariants which are independent and symmetric functions of the principal stretches, some of which may have useful symmetry built into them (see, e.g., Criscione et al. (2000) and Ennis and Kindmann, 2006).

At a formal level, one can, in principle, reformulate any strain-energy function into any such complete set of invariants or stretches. The choice of one set or another only becomes important when either trying to described some basic intrinsic properties of the system that are better captured with a given set, or when approximating a strain-energy function within a given space of functions. The question is then: what is the choice of invariants that best captures the behaviour of a subject material? Here, we provide a systematic method to obtain invariants that generalise the classical ones and that can be tuned to match pseudo-universal relationships in simple deformations, independently of the specific form of the strain-energy function.

2. Generalised invariants

If we are agnostic about the choice of invariants, we can introduce a set of invariants that depend on one parameter and choose the parameter judiciously when needed. Therefore, to bridge between the description in terms of $\{I_1, I_2, I_3\}$ and $\{i_1, i_2, i_3\}$, we introduce the set:

$$\mathcal{J}_\alpha = \lambda_1^\alpha + \lambda_2^\alpha + \lambda_3^\alpha, \quad (8)$$

$$\mathcal{J}_{-\alpha} = \lambda_1^{-\alpha} + \lambda_2^{-\alpha} + \lambda_3^{-\alpha}, \quad (9)$$

$$\mathcal{K}_\alpha = \lambda_1^\alpha \lambda_2^\alpha \lambda_3^\alpha, \quad (10)$$

where $\alpha \neq 0$. It should be clear that $\{\mathcal{J}_\alpha, \mathcal{J}_{-\alpha}, \mathcal{K}_\alpha\}$ forms a complete set and that the invariants $\{I_1, I_2, I_3\}$ and $\{i_1, i_2, i_3\}$ are recovered for $\alpha = 2$ and $\alpha = 1$, respectively.

We note that the generalised invariant \mathcal{J}_α has implicitly been used in the literature. For instance, the celebrated model of Ogden (1972):

$$W = \sum_{j=1}^n \frac{\mu_j}{\alpha_j} (\lambda_1^{\alpha_j} + \lambda_2^{\alpha_j} + \lambda_3^{\alpha_j} - 3) , \quad (11)$$

can be written as:

$$W = \sum_{j=1}^n \frac{\mu_j}{\alpha_j} (\mathcal{J}_{\alpha_j} - 3). \quad (12)$$

In Ogden (1972), a satisfactory agreement with the data for incompressible natural rubber was obtained on using $n = 3$, with $\alpha_1 = 1.3$, $\alpha_2 = 5$ and $\alpha_3 = -2$.¹ Similarly, in a recent work by Anssari-Benam (2023), a non-separable *parent* (principal) stretches-based model was proposed of the form:

$$W = \sum_{j=1}^n \frac{3(n_j - 1)}{2n_j} \mu_j N_j \left[\frac{1}{3N_j(n_j - 1)} (\lambda_1^{\alpha_j} + \lambda_2^{\alpha_j} + \lambda_3^{\alpha_j} - 3) - \ln \left(\frac{\lambda_1^{\alpha_j} + \lambda_2^{\alpha_j} + \lambda_3^{\alpha_j} - 3N_j}{3 - 3N_j} \right) \right], \quad (13)$$

where $\lambda_1 \lambda_2 \lambda_3 = 1$ to enforce the condition of incompressibility, μ_i are stress-like, and N_i , n_i , and α_i are dimensionless, model parameters subjected to n_i , $N_i \in \mathbb{R}^+$, and $\alpha_i \in \mathbb{R}$. With $j = 1$, this model recovers the one-term Ogden model when $N \rightarrow \infty$. It is also parent to other models found in the literature, including the generalised Gent (1996) model (proposed by Murphy, 2006) when $n \rightarrow \infty$ (the special case $\alpha = 2$ recovers exactly the Gent model).

Thus far we have merely stipulated that we can reformulate a strain-energy function W in terms of different deformation variables, including the generalised set of invariants in Eqs. (8) to (10). The advantage of using one or another only becomes clear when we consider *reduced models*.

3. Reduced models

We restrict our attention to the particular but important case of incompressible isotropic elasticity. In this case, the three invariants $\{I_1, I_2, I_3\}$ can be rewritten as:

$$I_1 = \lambda_1^2 + \lambda_2^2 + \lambda_3^2, \quad I_2 = \lambda_1^{-2} + \lambda_2^{-2} + \lambda_3^{-2}, \quad (14)$$

with $I_3 = \lambda_1^2 \lambda_2^2 \lambda_3^2 = 1$. Any strain-energy function W expressed in terms of the (principal) invariants; i.e., $W_{\text{inv}} = W_{\text{inv}}(I_1, I_2)$ where the subscript ‘inv’ denotes invariants, may also be easily expressed in the form $W_{\text{ps}} = W_{\text{ps}}(\lambda_1, \lambda_2, \lambda_3)$, and conversely, any $W = W_{\text{inv}}(\lambda_1, \lambda_2, \lambda_3)$ function enjoying symmetry may be re-written in terms of I_1 and I_2 using the relationship given by Rivlin and Sawyers (1976):

$$\lambda_i = \frac{1}{\sqrt{3}} \sqrt{I_1 + 2\sqrt{(I_1^2 - 3I_2)} \cos \left[\frac{1}{3} (\varphi + 2\pi i) \right]}, \quad i = 1, 2, 3, \quad (15)$$

where:

$$\varphi = \cos^{-1} \left[\frac{2I_1^3 - 9I_1 I_2 + 27}{2(I_1^2 - 3I_2)^{3/2}} \right]. \quad (16)$$

Since every complete set of invariants is always in a one-to-one correspondence with the principal stretches, a similar equivalent representation for any given set is always possible. This equivalence, however, is only valid when we consider a *complete* set of invariants. It is, often convenient to consider strain-energy functions that depend on a *reduced* set of invariants. In that case, the choice of the invariant(s) becomes important, and the ensuing mathematical and modelling implications of the considered class of models in capturing the true deformation of a given material must be analysed and understood.

¹We note that only two of the three invariants used there are independent in the isochoric space. This means that there may be some redundancy in the fitting procedure therein.

A paradigm of this reduced functional dependency may perhaps be best represented by the so called *generalised neo-Hookean*, or *first-invariant*, materials with the strain-energy function given as a function of I_1 only, that is $W = W(I_1)$. Another class of models (and in some sense complementary to the foregoing) are the *second-invariant models* recently proposed by Kuhl and Goriely (2024), which depend exclusively on the second principal invariant I_2 counterpart, i.e., $W(I_2)$. These reduced models enjoy several attractive features. For example, first-invariant models possess a certain mathematical simplicity since they are easy to apply and implement when modelling a single stress or strain component, as is the case in uniaxial extension or simple shear deformations. For such deformations, they provide, at least to a first approximation, a close correlation with the data. Additionally, they may also be connected with some structural features, e.g., statistical average-stretch network models have a strain energy function of this form (Beatty, 2003). Similarly, the second-invariant models have a certain mathematical simplicity while playing a role in better understanding the complex material behaviours such as those exhibited by soft biological tissues (see Kuhl and Goriely, 2024). However, it is also well known that when modelling the three dimensional deformation of soft materials, such as in the case of biaxial deformation or when analysing the normal stresses accompanying shear, the fact that either of the invariants is missing is a clear deficiency; a problem shared with any such model with an incomplete dependency on the full set of invariants (see, e.g., Horgan and Saccomandi, 1999; Saccomandi, 2001; Wineman, 2005; and Horgan and Smayda, 2012).

A new class of reduced models may be constructed by using the general invariants given in Eqs. (8) to (10). Before reduction, the general form of a model based on those general invariants is:

$$W = W(\mathcal{J}_\alpha, \mathcal{J}_{-\alpha}, \mathcal{K}_\alpha). \quad (17)$$

For incompressible materials, $\mathcal{K}_\alpha = 1$, and we restrict our attention to models that depend only on \mathcal{J}_α to define *general-invariant models* as those that only depend on \mathcal{J}_α :

$$W = W(\mathcal{J}_\alpha). \quad (18)$$

We see that these *general-invariant models* bridge the first- and second-invariant models which are obtained by fixing $\alpha = 2$, and $\alpha = -2$, respectively. Here, α is a constitutive parameter which remains to be determined from the experimental data so that the model suitably captures the deformation behaviour of a given material. We observe that the one-term form of the Ogden model in Eq. (11); $W = \mu(\mathcal{J}_\alpha - 3)/\alpha$, and the one-term expansion of the parent model in Eq. (13) of the form:

$$W = \frac{3(n-1)}{2n} \mu N \left[\frac{1}{3N(n-1)} (\mathcal{J}_\alpha - 3) - \ln \left(\frac{\mathcal{J}_\alpha - 3N}{3 - 3N} \right) \right], \quad (19)$$

both belong to the class of general-invariant models.

Separate from the geometric interpretation of the classical principal invariants I_1, I_2 and I_3 (Kearsley, 1989), the appearance of these invariants in a strain-energy function is often a result of particular kinematic assumptions. For example, in average chain models, the existence of I_1 is based on the assumption of the affine deformation of chains (Beatty, 2003), and I_2 stems from the consideration of a topological tube constraint around the chains (Fried, 2002; Khiêm and Itskov, 2016; Anssari-Benam et al., 2021). However, both these assumptions are mathematical idealisation, and in reality, the mechanics of the deformation of the chains may not follow either of the assumed kinematic scenarios. This possibility further justifies the introduction of the generalised invariant \mathcal{J}_α . Therefore, the general-invariant strain-energy function $W(\mathcal{J}_\alpha)$ significantly generalises the classical models that are based on only a single invariant, opening a new horizon with great potential for consideration of this type of constitutive equations.

A challenge, however, that arises when considering α as a constitutive parameter is that the fitting of such a parameter is highly nonlinear and as such it may introduce the problems discussed by Ogden et al. (2004), including the non-uniqueness of the optimal set of parameters and issues related to the convergence of the numerical methods. These problems may be overcome by a two-step process where we first obtain the exponent α and then a functional form of $W(\mathcal{J})$. The key in this task is the existence of pseudo-universal relationships, independent of W . Using these relationships, the first step is to quantify the exponent α from basic experimental deformation data, independently from the choice of W . Once the exponent is known, the second step is to use classical methods to fit data with the *generalised-invariant model*.

4. Pseudo-universal relationships

For incompressible isotropic materials, the representation formula linking the Cauchy stress tensor \mathbf{T} to the strain-energy function reads:

$$\mathbf{T} = -p\mathbf{I} + 2W_1\mathbf{B} - 2W_2\mathbf{B}^{-1}, \quad (20)$$

where W_1 and W_2 are the partial derivatives of the classical principal invariants-based model, $W(I_1, I_2)$, with respect to I_1 and I_2 , respectively. In terms of \mathcal{J} and using the chain rule, the representation formula may be recast as:

$$\mathbf{T} = -p\mathbf{I} + 2W' \left(\frac{\partial \mathcal{J}}{\partial I_1} \mathbf{B} - \frac{\partial \mathcal{J}}{\partial I_2} \mathbf{B}^{-1} \right), \quad (21)$$

with the prime denoting the partial derivative of $W(\mathcal{J})$ with respect to \mathcal{J} , and where for simplicity, we have dropped the dependency notation of \mathcal{J} on α .

Using the incompressibility condition we can write the invariant \mathcal{J} as:

$$\mathcal{J} = \lambda_1^\alpha + \lambda_2^\alpha + \frac{1}{\lambda_1^\alpha \lambda_2^\alpha}, \quad (22)$$

from which, we obtain:

$$\begin{cases} \frac{\partial \mathcal{J}}{\partial I_1} = -\frac{\alpha \lambda_1^2 \lambda_2^2 [\lambda_1^4 \lambda_2^{\alpha+4} - \lambda_2^4 \lambda_1^{\alpha+4} + \lambda_1^{\alpha+2} - \lambda_2^{\alpha+2} - \lambda_1^{-\alpha} \lambda_2^{-\alpha} (\lambda_1^2 - \lambda_2^2)]}{2(\lambda_1^4 \lambda_2^2 - 1)(\lambda_1^2 \lambda_2^4 - 1)(\lambda_1^2 - \lambda_2^2)}, \\ \frac{\partial \mathcal{J}}{\partial I_2} = \frac{\alpha \lambda_1^2 \lambda_2^2 [\lambda_1^4 \lambda_2^{\alpha+2} - \lambda_2^4 \lambda_1^{\alpha+2} - \lambda_1^{-\alpha+2} \lambda_2^{-\alpha+2} (\lambda_1^2 - \lambda_2^2) + \lambda_1^\alpha - \lambda_2^\alpha]}{2(\lambda_1^4 \lambda_2^2 - 1)(\lambda_1^2 \lambda_2^4 - 1)(\lambda_1^2 - \lambda_2^2)}, \end{cases} \quad (23)$$

subject to the condition:

$$(\lambda_1^4 \lambda_2^2 - 1)(\lambda_1^2 \lambda_2^4 - 1)(\lambda_1^2 - \lambda_2^2) \neq 0. \quad (24)$$

From the (23) we see that when $\alpha = 2$ we have $\partial \mathcal{J} / \partial I_2 = 0$, and when $\alpha = -2$ we get $\partial \mathcal{J} / \partial I_1 = 0$, as expected. The special cases when (24) is not satisfied will be discussed later.

The next step is to specialise the representation formula in (21), using the relationships in (23), for the particular in-plane deformations regularly employed in experiments to characterise the mechanical behaviour of soft materials (see Mihai and Goriely (2017) for an overview).

4.1. General biaxial deformation

An incompressible *biaxial deformation* is a homogeneous deformation defined by the following kinematics:

$$x_1 = \lambda_1 X_1, \quad x_2 = \lambda_2 X_2, \quad x_3 = (\lambda_1 \lambda_2)^{-1} X_3. \quad (25)$$

Accordingly, using the representation formula in (21), the Cauchy stress is diagonal, $\mathbf{T} = \text{diag}(T_1, T_2, T_3)$. With the assumption of plane stress $T_3 = 0$, the remaining principal components of the Cauchy stress are:

$$T_1 = 2W'(\mathcal{J}) \left[\frac{\partial \mathcal{J}}{\partial I_1} (\lambda_1^2 - \lambda_1^{-2} \lambda_2^{-2}) - \frac{\partial \mathcal{J}}{\partial I_2} (\lambda_1^{-2} - \lambda_1^2 \lambda_2^2) \right], \quad (26)$$

$$T_2 = 2W'(\mathcal{J}) \left[\frac{\partial \mathcal{J}}{\partial I_1} (\lambda_2^2 - \lambda_1^{-2} \lambda_2^{-2}) - \frac{\partial \mathcal{J}}{\partial I_2} (\lambda_2^{-2} - \lambda_1^2 \lambda_2^2) \right]. \quad (27)$$

By taking the ratio of these two equations, we can eliminate the dependence on the strain-energy function W to obtain the *pseudo-universal relationship*:

$$\mathcal{R}(\alpha) := \frac{T_1}{T_2} - \frac{\frac{\partial \mathcal{J}}{\partial I_1}(\lambda_1^2 - \lambda_1^{-2} \lambda_2^{-2}) - \frac{\partial \mathcal{J}}{\partial I_2}(\lambda_1^{-2} - \lambda_1^2 \lambda_2^2)}{\frac{\partial \mathcal{J}}{\partial I_1}(\lambda_2^2 - \lambda_1^{-2} \lambda_2^{-2}) - \frac{\partial \mathcal{J}}{\partial I_2}(\lambda_2^{-2} - \lambda_1^2 \lambda_2^2)} = 0. \quad (28)$$

This relationship, while independent of the choice of W , is a function of the constitutive parameter α . Therefore, it is a *pseudo-universal* relationship rather than a *universal* relationship. In other words, for any fixed value of α , Eq. (28) is a universal relationship. For instance, the case $\mathcal{R}(2)$ and $\mathcal{R}(-2)$ have been previously described by Destrade et al. (2017) and Kuhl and Goriely (2024), respectively. Universal relationships are usually determined from coaxiality of \mathbf{T} and \mathbf{B} ; i.e., $\mathbf{T}\mathbf{B} = \mathbf{B}\mathbf{T}$. The pseudo-universal relationship $\mathcal{R}(\alpha)$, however, was obtained by considering the principal stresses and stretches following a general method first described by Pucci and Saccomandi (1997); see also Saccomandi and Vergori (2019) for the application to other classes of materials.

The substitution of the relationships in Eq. (23) into Eq. (28) leads to the following explicit pseudo-universal relationship for general biaxial deformations:

$$\mathcal{R}(\alpha) = \frac{(\lambda_1^4 \lambda_2^4 - \lambda_1^2 \lambda_2^6 + \lambda_1^{2\alpha+2} \lambda_2^\alpha + \lambda_1^{2\alpha+2} \lambda_2^{\alpha+6} - \lambda_1^{2\alpha} \lambda_2^{\alpha+2} - \lambda_1^{2\alpha+4} \lambda_2^{\alpha+4} - \lambda_1^2 + \lambda_2^2)(\lambda_1^4 \lambda_2^2 - 1)}{(-\lambda_1^{\alpha+6} \lambda_2^{2\alpha+2} + \lambda_1^{\alpha+4} \lambda_2^{2\alpha+4} + \lambda_1^6 \lambda_2^2 - \lambda_1^4 \lambda_2^4 + \lambda_1^{\alpha+2} \lambda_2^{2\alpha} - \lambda_1^\alpha \lambda_2^{2\alpha+2} - \lambda_1^2 + \lambda_2^2)(\lambda_1^2 \lambda_2^4 - 1)}. \quad (29)$$

This expression may seem lengthy but it can easily be utilised, since the values of λ_1 and λ_2 are known from experiments and therefore the computation of α is achieved by the standard correlation method.

Now let us recall the condition (24). One of the scenarios in which this condition is violated is under equi-biaxial deformation ($\lambda_1 = \lambda_2$). In the limit $\lambda_1 \rightarrow \lambda_2$, we have $T_1 \rightarrow T_2$ and the relationship in (28) is identically satisfied. Therefore, in this case, the pseudo-universal relationship (28) does not provide any information on the exponent α . Another case for which the condition (24) is not satisfied is under uniaxial deformation, with the following kinematics:

$$x_1 = \lambda X_1, \quad x_2 = \frac{1}{\sqrt{\lambda}} X_2, \quad x_3 = \frac{1}{\sqrt{\lambda}} X_3. \quad (30)$$

Again, the pseudo-universal relationship (28) does not provide any information about α , since the only non-zero component of stress is T_1 . Hence, we conclude that uniaxial data cannot be used to fix the exponent α through the pseudo-universal relationship.

4.2. Pure shear deformation

Another typical deformation used in characterising the mechanical behaviour of rubber-like materials is pure shear:

$$\lambda_1 = \lambda, \quad \lambda_2 = 1, \quad \lambda_3 = \lambda^{-1}. \quad (31)$$

In this case, (28) simplifies to:

$$\mathcal{R}(\alpha) := \frac{T_1}{T_2} - 1 - \lambda^\alpha = 0. \quad (32)$$

This is a conveniently simple and compact relationship that can be used together with experimental pure shear data to determine the value of α . Note that we can re-write this last expression as:

$$\ln \left(\frac{T_1}{T_2} - 1 \right) = \alpha \ln(\lambda), \quad (33)$$

which gives a simple linear relationship to fit between $\ln[(T_1/T_2 - 1)]$ and $\ln \lambda$.

4.3. Simple shear deformation

Simple shear deformation is another classical test used to characterise the mechanical behaviour of soft solids. It is given by:

$$x_1 = X_1 + \kappa X_2, \quad x_2 = X_2, \quad x_3 = X_3, \quad (34)$$

where κ quantifies the amount of shear. Subject to the boundary condition $T_{33} = 0$, the remaining components of the Cauchy stress tensor under simple shear are:

$$\begin{cases} T_{11} = \frac{\partial \mathcal{J}}{\partial I_1} W'(\mathcal{J}) \kappa^2, \\ T_{22} = -\frac{\partial \mathcal{J}}{\partial I_2} W'(\mathcal{J}) \kappa^2, \\ T_{12} = \left(\frac{\partial \mathcal{J}}{\partial I_1} + \frac{\partial \mathcal{J}}{\partial I_2} \right) W'(\mathcal{J}) \kappa, \end{cases} \quad (35)$$

where we note $I_1 = I_2 = 3 + \kappa^2$. From these relationships, Rivlin's classical universal relationship $T_{11} - T_{22} = \kappa T_{12}$ is obtained, which generalises to the new pseudo-universal relationship as follows:

$$\mathcal{R}(\alpha) := \frac{T_{11}}{T_{22}} + \frac{\partial \mathcal{J} / \partial I_1}{\partial \mathcal{J} / \partial I_2} = 0, \quad (36)$$

with $\partial \mathcal{J} / \partial I_1$ and $\partial \mathcal{J} / \partial I_2$ given by (23). Further, in simple shear deformation, the principal stretches λ_1 and λ_2 are directly related to κ :

$$\lambda_1 = \frac{\kappa}{2} + \sqrt{1 + \frac{\kappa^2}{4}}, \quad \lambda_2 = \lambda_1^{-1} = -\frac{\kappa}{2} + \sqrt{1 + \frac{\kappa^2}{4}}, \quad \lambda_3 = 1. \quad (37)$$

Hence, after simplification, we obtain:

$$\mathcal{R}(\alpha) = \frac{T_{11}}{T_{22}} + \frac{\left(\kappa - \sqrt{\kappa^2 + 4} \right) \left(-\frac{\kappa}{2} + \frac{\sqrt{\kappa^2 + 4}}{2} \right)^\alpha - \left(\kappa + \sqrt{\kappa^2 + 4} \right) \left(\frac{\kappa}{2} + \frac{\sqrt{\kappa^2 + 4}}{2} \right)^\alpha + 2\sqrt{\kappa^2 + 4}}{\left(\kappa + \sqrt{\kappa^2 + 4} \right) \left(-\frac{\kappa}{2} + \frac{\sqrt{\kappa^2 + 4}}{2} \right)^\alpha - \left(\kappa - \sqrt{\kappa^2 + 4} \right) \left(\frac{\kappa}{2} + \frac{\sqrt{\kappa^2 + 4}}{2} \right)^\alpha - 2\sqrt{\kappa^2 + 4}}. \quad (38)$$

This pseudo-universal relationship depends only on α , as κ is measured or controlled in experiments. Therefore, by correlating this relationship with the data $\{T_{11}/T_{22}, \kappa\}$, it is straightforward to obtain α . However, we note that in practice, it may be difficult to obtain T_{11} and T_{22} experimentally. It is also observed from (38) that when $\alpha = 2$, i.e., in the case of a first-invariant material, we obtain the well-known universal result $T_{22} = 0$, as noted by Destrade et al. (2015) in the context of modelling brain tissue shearing.

4.4. Simple torsion deformation

The previous cases all belonged to the class of homogeneous deformations. It is also possible to derive such pseudo-universal relationships for inhomogeneous deformations. A typical example is the case of simple torsion of a long circular cylinder of radius R_0 :

$$r = R, \quad \theta = \Theta + \tau Z, \quad z = Z, \quad (39)$$

where τ is the twist per unit length. From the equilibrium condition $\text{div } \mathbf{T} = 0$ in direction \mathbf{e}_r , we obtain the pressure as:

$$p(R) = \tau^2 \int_R^{R_0} r W'(\mathcal{J}) \frac{\partial \mathcal{J}}{\partial I_1} dr. \quad (40)$$

The non-zero stress components will therefore be given by:

$$\begin{cases} T_{rr} = -p, \\ T_{\theta\theta} = -2p + 2\tau^2 R^2 W'(\mathcal{J}) \frac{\partial \mathcal{J}}{\partial I_1}, \\ T_{zz} = -2p - 2\tau^2 R^2 W'(\mathcal{J}) \frac{\partial \mathcal{J}}{\partial I_2}, \\ T_{z\theta} = 2\tau R W'(\mathcal{J}) \left(\frac{\partial \mathcal{J}}{\partial I_1} + \frac{\partial \mathcal{J}}{\partial I_2} \right). \end{cases} \quad (41)$$

The usual universal relationship related to the coaxiality of \mathbf{T} and \mathbf{B} holds: $T_{\theta\theta} - T_{zz} = \tau R T_{\theta z}$. By using the components of the Cauchy stress in (41), we obtain a new pseudo-universal relationship as:

$$\frac{\partial \mathcal{J}}{\partial I_2} (T_{\theta\theta} - 2T_{rr}) + \frac{\partial \mathcal{J}}{\partial I_1} (T_{zz} - 2T_{rr}) = 0. \quad (42)$$

Again, this relation is independent of the choice of the strain-energy function W as it only depends on α . However, in practice, this relationship is not helpful since stresses cannot be easily measured. Instead, in a typical experiment, we have access to the total moment \mathcal{M} and the total axial force \mathcal{N} , given by:

$$\begin{cases} \mathcal{M} = 4\pi\tau \int_0^{R_0} W'(\mathcal{J}) \left(\frac{\partial \mathcal{J}}{\partial I_1} + \frac{\partial \mathcal{J}}{\partial I_2} \right) R^3 dR, \\ \mathcal{N} = -4\pi\tau^2 \int_0^{R_0} W'(\mathcal{J}) \left(\frac{1}{2} \frac{\partial \mathcal{J}}{\partial I_1} + \frac{\partial \mathcal{J}}{\partial I_2} \right) R^3 dR. \end{cases} \quad (43)$$

Note that for the simple torsion deformation, the principal stretches λ_i are given by (Horgan and Murphy, 2023):

$$\lambda_1 = \frac{1}{2} \left(k + \sqrt{4 + k^2} \right), \quad \lambda_2 = \lambda_1^{-1} = \frac{1}{2} \left(-k + \sqrt{4 + k^2} \right), \quad \lambda_3 = 1, \quad (44)$$

where $k = R\tau$. It follows, after some manipulation and simplification, that:

$$\frac{\partial \mathcal{J}}{\partial I_1} + \frac{\partial \mathcal{J}}{\partial I_2} = -\alpha \frac{\left[\left(-\frac{k}{2} + \frac{\sqrt{k^2 + 4}}{2} \right)^\alpha - \left(\frac{k}{2} + \frac{\sqrt{k^2 + 4}}{2} \right)^\alpha \right]}{2k\sqrt{k^2 + 4}}, \quad (45)$$

and:

$$\begin{aligned} \frac{1}{2} \frac{\partial \mathcal{J}}{\partial I_1} + \frac{\partial \mathcal{J}}{\partial I_2} = & -\frac{3\alpha}{8k^2\sqrt{k^2 + 4}} \left[-\frac{2}{3}\sqrt{k^2 + 4} + \left(k + \frac{\sqrt{k^2 + 4}}{3} \right) \left(-\frac{k}{2} + \frac{\sqrt{k^2 + 4}}{2} \right)^\alpha \right. \\ & \left. - \left(k - \frac{\sqrt{k^2 + 4}}{3} \right) \left(\frac{k}{2} + \frac{\sqrt{k^2 + 4}}{2} \right)^\alpha \right]. \end{aligned} \quad (46)$$

These two relationships are clearly only a function of α , since k is experimentally known. Therefore, one possibility to obtain a pseudo-universal relationship involving α is by taking the ratio of \mathcal{M} and \mathcal{N} , in which case W' is eliminated. Accordingly, if we consider the one-term Ogden model $W = \mu(\mathcal{J} - 3)/\alpha$ we get:

$$\begin{cases} \mathcal{M} = 4\pi\tau \frac{\mu}{\alpha} \int_0^{R_0} \left(\frac{\partial \mathcal{J}}{\partial I_1} + \frac{\partial \mathcal{J}}{\partial I_2} \right) R^3 dR, \\ \mathcal{N} = -4\pi\tau^2 \frac{\mu}{\alpha} \int_0^{R_0} \left(\frac{1}{2} \frac{\partial \mathcal{J}}{\partial I_1} + \frac{\partial \mathcal{J}}{\partial I_2} \right) R^3 dR. \end{cases} \quad (47)$$

By defining $\mathbb{I}(\alpha)$ as:

$$\mathbb{I}(\alpha) = \frac{\int_0^{R_0} \left(\frac{\partial \mathcal{J}}{\partial I_1} + \frac{\partial \mathcal{J}}{\partial I_2} \right) R^3 dR}{\int_0^{R_0} \left(\frac{1}{2} \frac{\partial \mathcal{J}}{\partial I_1} + \frac{\partial \mathcal{J}}{\partial I_2} \right) R^3 dR}, \quad (48)$$

and in view of the foregoing expressions for $\left(\frac{\partial \mathcal{J}}{\partial I_1} + \frac{\partial \mathcal{J}}{\partial I_2} \right)$ and $\left(\frac{1}{2} \frac{\partial \mathcal{J}}{\partial I_1} + \frac{\partial \mathcal{J}}{\partial I_2} \right)$, it is clear that \mathbb{I} will only be a function of α , and we thus arrive at the following special case of a pseudo-universal relationship:

$$\frac{\mathcal{M}}{\mathcal{N}} = -\frac{\mathbb{I}}{\tau}. \quad (49)$$

This relationship is a pseudo-universal one since it only depends on α ; however, only for the considered case of the one-term Ogden model. We remark that we have not yet found other strain-energy functions to deduce a pseudo-universal relationship from the global quantities \mathcal{M} and \mathcal{N} .

5. A two-step fitting method

We can now use the pseudo-universal relationships presented in Section 4 to obtain a suitable strain-energy function W that only depends on a single invariant. We assume that we have a suitable set of data, that is either biaxial, pure shear, or simple torsion data. The goal is to find a strain-energy function $W(\mathcal{J}_\alpha)$ that best fits with data. To do this, we first need to identify the exponent α and then the functional form $W(\mathcal{J})$. We proceed in two steps:

First step: the exponent. Given the dataset, we create the combination of strains and stresses that appear in the corresponding pseudo-universal relationship, and denote them symbolically as the strain data $\mathbf{x} = \{x_i, i = 1, \dots, N\}$ and stress data $\mathbf{y} = \{y_i, i = 1, \dots, N\}$. For each point we compute the residual $r_i = \mathcal{R}(x_i, y_i; \alpha)$. Then, we find α by minimising $R = \sum_{i=1}^N r_i^2$. The result of this step is the exponent that best captures the universal character of the data, *independently of the functional choice of the strain-energy function*.

We note that this step can be used to check whether a first- or second-invariant model is suitable. Indeed, by obtaining the exponent α via fitting the pseudo-universal relationships against the related experimental data, we can identify whether a pre-assumed first-invariant model $W(I_1)$ or a second invariant-only counterpart $W(I_2)$ model is the correct class of functions to describe the deformation of the given specimen. If the best fits of the pseudo-universal relationships to the data are obtained via values of α is close to 2 or -2, then the first- or second- invariant models may be deemed appropriate for those datasets.

Second step: the strain-energy function. Once the exponent α is known, we can fix it and fit the strain energy function $W(\mathcal{J})$ to the full deformation data. An advantage is that the function W now does not depend on α , which greatly simplifies the fitting process and for which there are plenty of methods.

This two-step process not only facilitates a better likelihood of achieving a unique fit by fixing one parameter that contributes to the nonlinearity of the optimisation (as the exponent of λ); see Ogden et al. (2004) for a detailed elaboration on the sources of non-uniqueness in problems of nonlinear optimisation, but also streamlines the process of fitting the model to the deformation data by reducing the number of model parameters left to be identified.

5.1. First step: finding the generalised invariant for experimental data

Next we consider the datasets of Jones and Treloar (1975) and Fukahori and Seki (1992) that provide pure shear deformation data, and Kawabata et al. (1981) for both biaxial and pure shear deformations. For the fitting process of W , we will use the one-term expansion of the model by Anssari-Benam (2023) given in Eq. (19), and that of the Ogden model.

5.1.1. Example 1: pure shear data

The first example is based on data from Jones and Treloar (1975), where they report on the two components of stress, in our notation T_1 and T_2 , in pure shear deformation of samples made of a rubber material (see Table A1).

On fitting the pseudo-universal relationship (33) to the data, we easily obtain the exponent α for these specimens. The plot in Fig. 1 demonstrates the result, with the obtained value of $\alpha = 2.61$ (all numerical values are given to two decimal places). For comparison, we also show in Fig. 1 the prediction given by when $\alpha = 2$; i.e., a generalised neo-Hookean model. That prediction is given by a dotted red line. Notable deviations from the data are observed at both small and higher ranges of deformation when $\alpha = 2$. The choice $\alpha = -2$ gives similarly poor results (not shown).

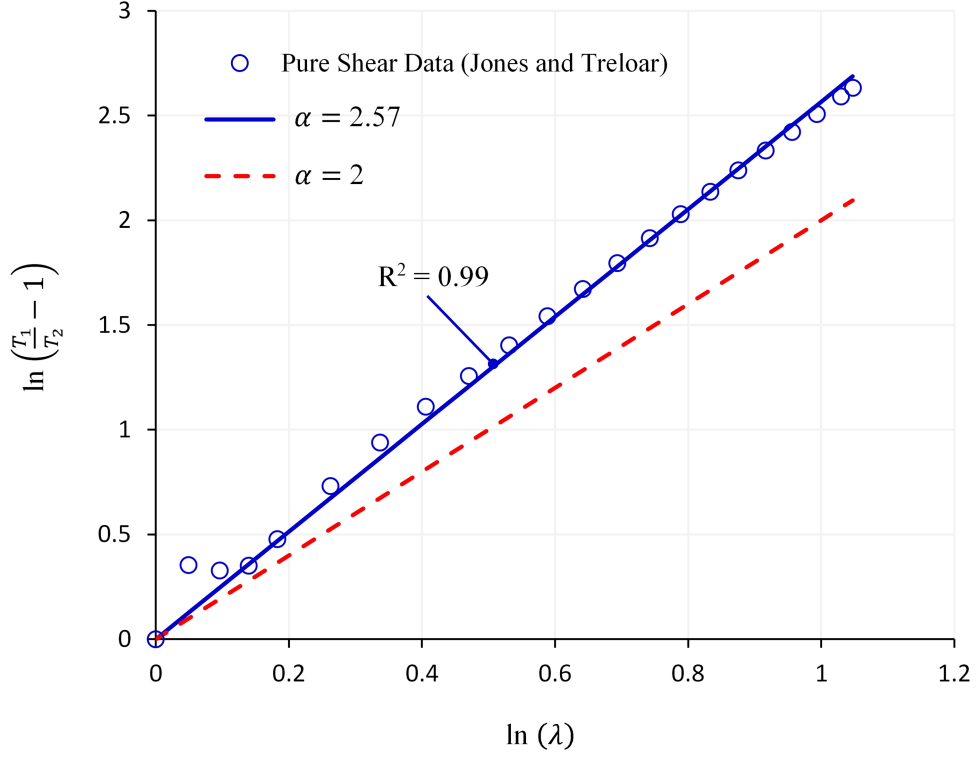


Fig. 1. Correlating the pseudo-universal relationship for pure shear in Eq. (33) with the data from Jones and Treloar (1975). The continuous blue line represents the best fit between the two with $\alpha = 2.61$. The dotted red line is the generalised neo-Hookean model; i.e., $\alpha = 2$. See the online version for the plots in colour.

Next, we consider the canonical dataset of Kawabata et al. (1981) for isoprene rubber vulcanizates (see the original work for the tabulated numerical datapoints). The fit of the pseudo-universal relationship in Eq. (33) to the pure shear deformation data of Kawabata et al. (1981) gives $\alpha = 1.31$ with R^2 value in excess of 0.99. The resulting correlation is shown in Fig. 2. Again, the generalised neo-Hookean model is observed to provide an inadequate fit.

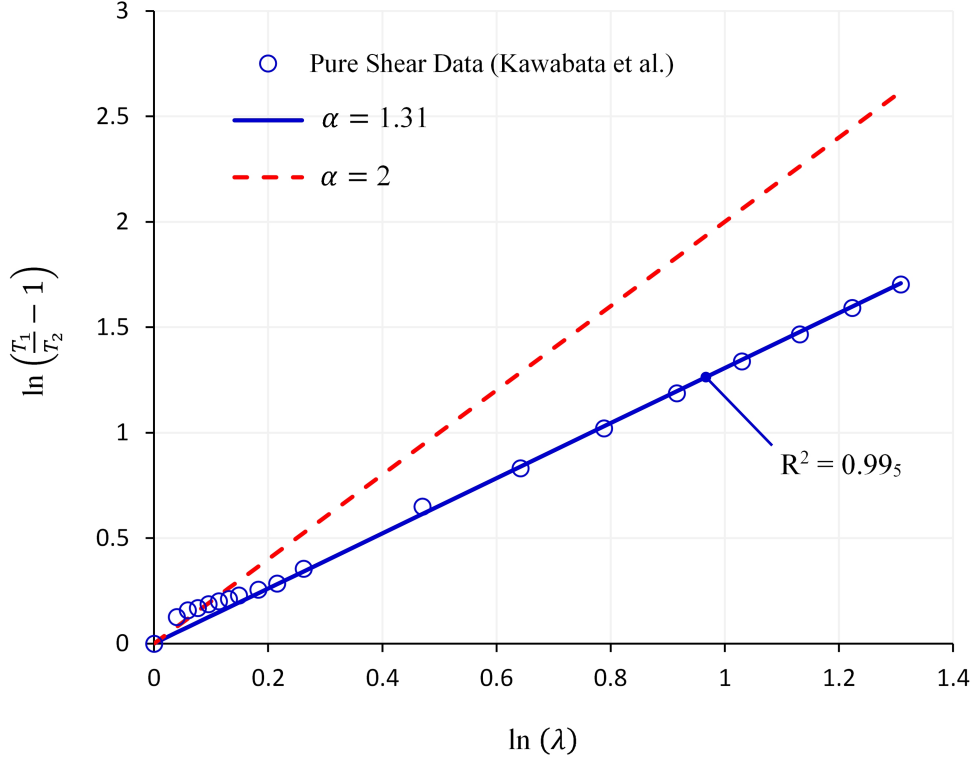


Fig. 2. Correlating the pseudo-universal relationship for pure shear in Eq. (33) with the data from Kawabata et al. (1981). The blue line represents the best fit ($\alpha = 1.31$). The dotted red line is the generalised neo-Hookean model ($\alpha = 2$). See the online version for the plots in colour.

The final example we consider here is the dataset due to Fukahori and Seki (1992), with the reported components of stress, T_1 and T_2 , in pure shear deformation of a carbon black reinforced natural rubber vulcanizate specimen (see Table A2). The plot in Fig. 3 shows the best fit between the pseudo-universal relationship (33) and the data, with the identified value of $\alpha = 1.88$. Again, the dotted red line in the plot is the prediction of the generalised neo-Hookean model. We note that while a good fit is obtained with the data at smaller ranges of deformation when considering $\alpha = 2$, there is a notable deviation for larger deformations.

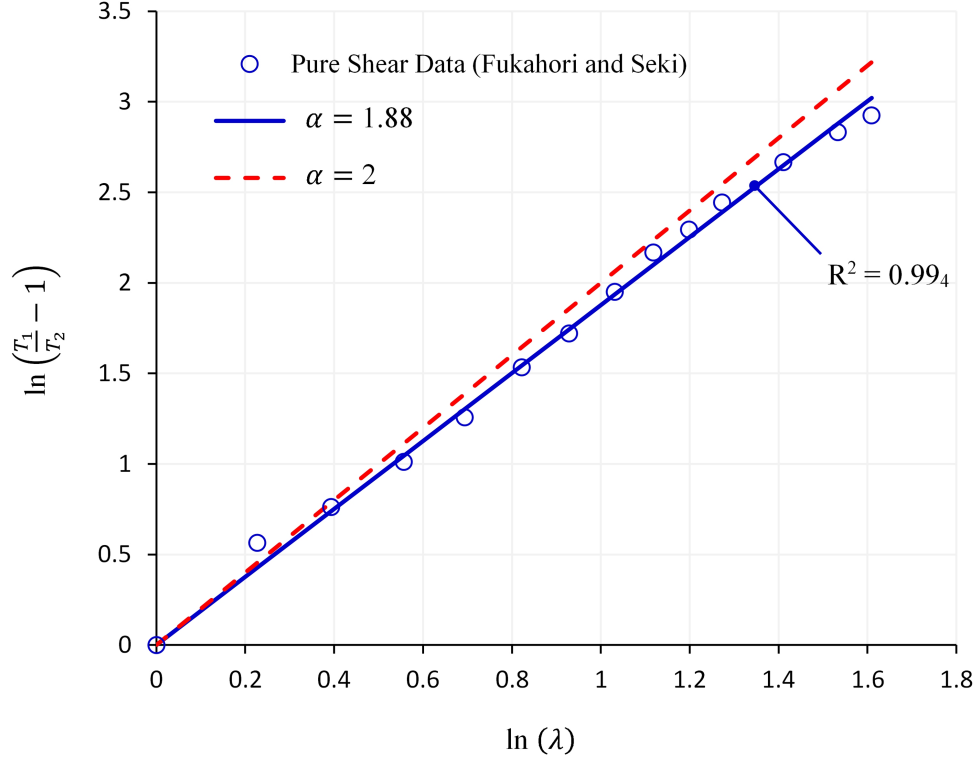


Fig. 3. Correlating the pseudo-universal relationship for pure shear in Eq. (33) with the data from Fukahori and Seki (1992). The continuous blue line is the best fit with $\alpha = 1.88$. The dotted red line is the generalised neo-Hookean model ($\alpha = 2$). See the online version for the plots in colour.

5.1.2. Example 2: biaxial data

To consider the application of the pseudo-universal relationship for biaxial deformation given by Eq. (29), we use the various biaxial deformation paths; i.e., various (λ_1, λ_2) pairs, given by Jones and Treloar (1975) and Kawabata et al. (1981). Starting with the former, we fit the pseudo-universal relationship (29) with the data for the four biaxial deformation paths provided in that study, namely for when $\lambda_2 = 1.502$, 1.984, 2.295 and 2.623, individually to each dataset (see Table A3 for the tabulated numerical datapoints). The results are shown in panels (a) to (d) of Fig. 4.

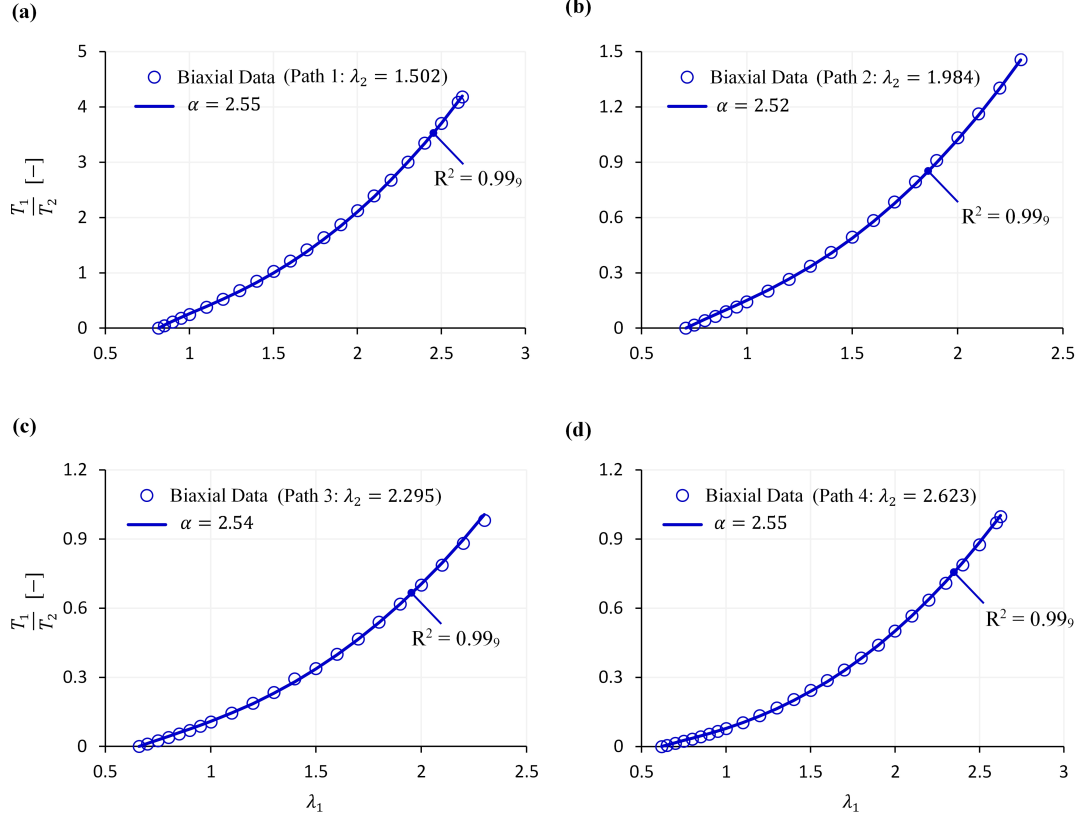


Fig. 4. Correlation between the pseudo-universal relationship for biaxial deformation in Eq. (29) and the data from Jones and Treloar (1975): Panels (a) to (d) represent the best fit for various biaxial deformation paths; $\lambda_2 = 1.502, 1.984, 2.295$ and 2.623 , respectively. Note that the axes scales are different in each panel.

It is interesting to note that even by fitting independently the four deformation paths to the pseudo-universal relationship for biaxial deformation in Eq. (29), the values of α are remarkably close to each other, only varying between 2.52 to 2.55. Moreover, recall that the identified value of α for these specimens on using the pseudo-universal relationship in Eq. (33) for pure shear deformation was also 2.57; see Fig. 1.

We next consider the biaxial deformation dataset of Kawabata et al. (1981). We choose three different deformation paths reported therein, and fit the pseudo-universal relationship (29) to the data for each path, individually. The results are shown in panels (a) to (c) of Fig. 5.

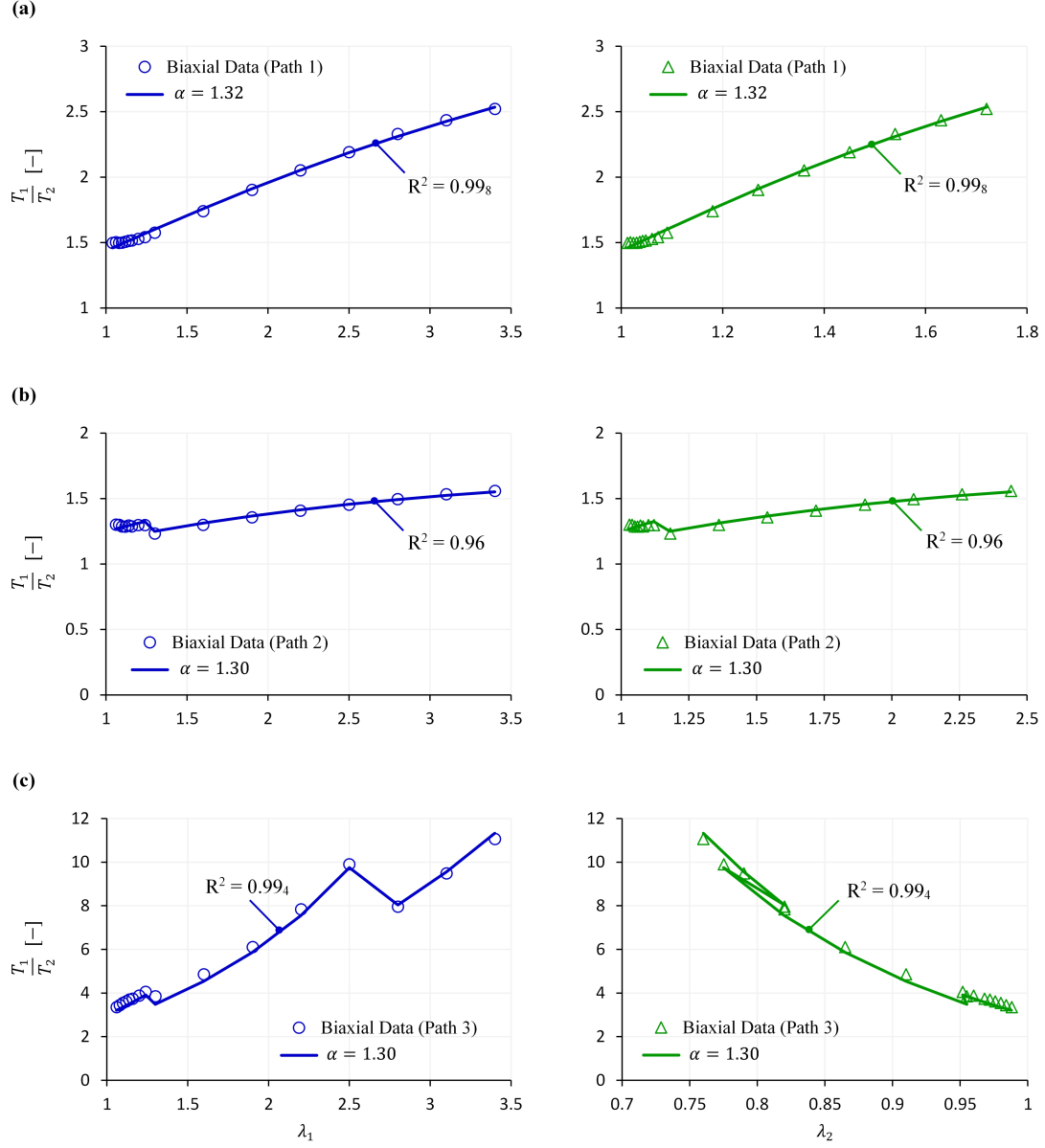


Fig. 5. Correlation between the pseudo-universal relationship for biaxial deformation in Eq. (29) and the data from Kawabata et al. (1981): Panels (a) to (c) represent the best fit for various biaxial deformation paths; 1, 2 and 3, respectively. Panels on the left are the plots in direction 1 (λ_1), while the right-hand side panels are those in direction 2 (λ_2). Note that the axes scales are different. See the online version for the plots in colour.

Similar to the observations regarding the biaxial data of Jones and Treloar (1975), we note again the remarkably close identified values of α here too, varying only between 1.30 to 1.32, recalling that the obtained value of α for these specimens on using the pseudo-universal relationship in Eq. (33) for pure shear deformation was also 1.31; see Fig. 2. These results suggest that the identified values of α in this approach are generic and represent some underlying intrinsic property of the material, as they are reproducibly found on using biaxial or pure shear deformations.

5.2. Second step: fitting the strain-energy function

The previous examples demonstrated how we can obtain the exponent α from data. Now that those exponents are fixed for each datasets, we can find a suitable functional form for the strain-energy function W to fit with the data. We focus on two possible choices: the classical one-term Ogden model and the parent model given by Eq. (19). We fit these two strain energy functions against the uniaxial deformation datasets of Jones and Treloar (1975), Kawabata et al. (1981) and Fukahori and Seki (1992), for which the values of exponent α were quantified in the previous section. We keep those values of α fixed in our fitting with the data here. Accordingly, for uniaxial deformation, the ensuing $T_{uni} - \lambda$ relationships of both models are:

$$\begin{cases} (T_{uni})^{og} = \mu \left(\lambda^\alpha - \frac{1}{\lambda^{\alpha/2}} \right), \\ T_{uni} = \frac{\mu \alpha}{2n} \frac{\lambda^\alpha + 2\lambda^{-\alpha/2} - 3nN}{\lambda^\alpha + 2\lambda^{-\alpha/2} - 3N} \left(\lambda^\alpha - \frac{1}{\lambda^{\alpha/2}} \right), \end{cases} \quad (50)$$

where the superscript ‘og’ in Eq. (50)₁ denotes the relationship pertaining to the Ogden model. These relationships are fitted with the uniaxial data of Jones and Treloar (1975), Kawabata et al. (1981) and Fukahori and Seki (1992) (data in Appendix A or in the original study). Note that for these datasets, we use values of $\alpha = 2.55, 1.88$ and 1.31 , respectively, as average values of α obtained from fitting the pseudo-universal relationships for biaxial and pure shear deformations with the respective datasets, presented in the previous section. The fitting results are shown in Fig. 6. The results highlight that: (i) the identified values of α using the pseudo-universal relationships provide a good fit with the data; and (ii) the functional choice of the model still plays an important role in obtaining a good fit, as the Ogden model provides notably less favourable fits.

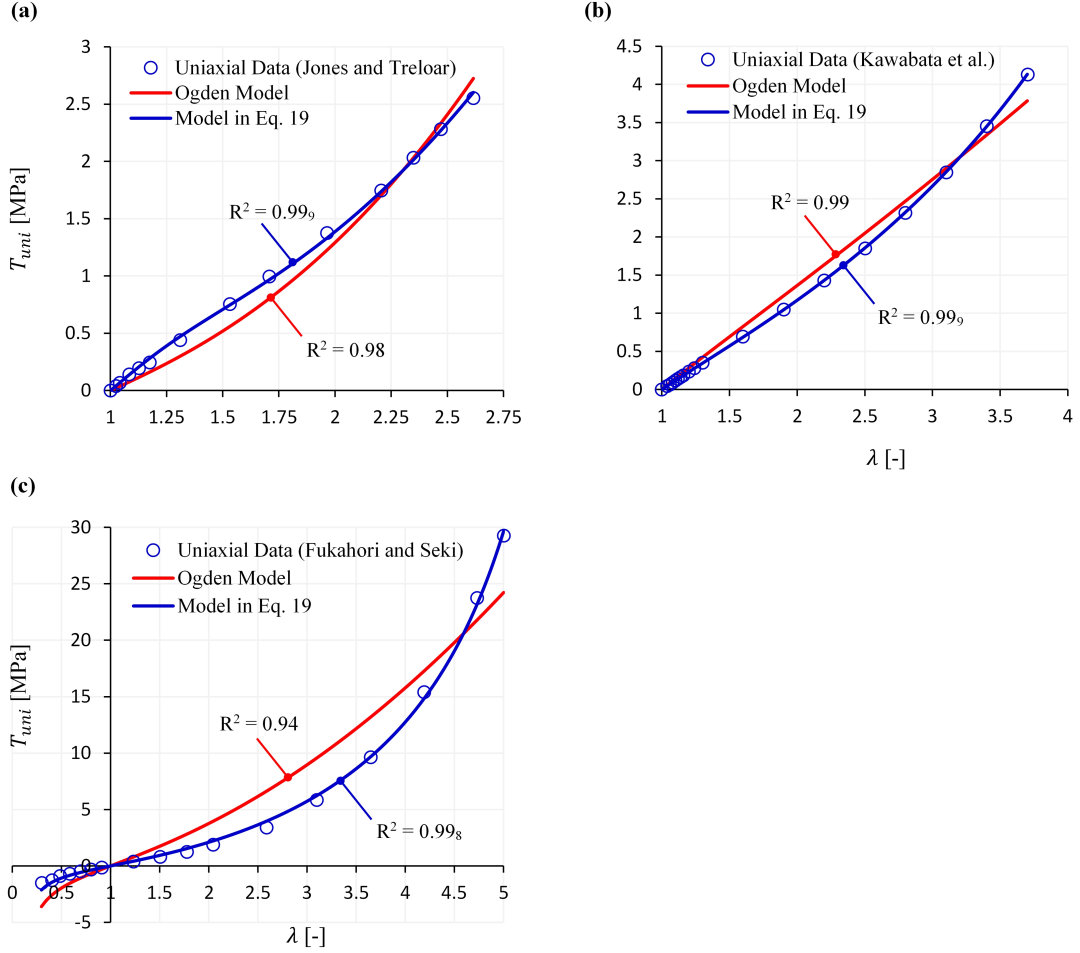


Fig. 6. Modelling results on fitting the Ogden model (red line) and that of Eq. (19) to the uniaxial deformation data of: (a) Jones and Treloar (1975) with the identified value of α using the pseudo-universal relationship as $\alpha = 2.55$ [-]. The obtained model parameter value(s) for the Ogden model is $\mu = 0.23$ [MPa], and for the model in Eq. (19) are: $\mu = 0.06$ [MPa], $N = 0.62$ [-] and $n = 0.35$ [-]; (b) Kawabata et al. (1981) with the identified value of α using the pseudo-universal relationship as $\alpha = 1.31$ [-]. The obtained model parameter value(s) for the Ogden model is $\mu = 0.74$ [MPa], and for the model in Eq. (19) are: $\mu = 0.77$ [MPa], $N = 4.74$ [-] and $n = 4.21$ [-]; and (c) Fukahori and Seki (1992) with the identified value of α using the pseudo-universal relationship as $\alpha = 1.88$ [-]. The obtained model parameter value(s) for the Ogden model is $\mu = 1.19$ [MPa], and for the model in Eq. (19) are: $\mu = 0.62$ [MPa], $N = 11.325$ [-] and $n = 19.18$ [-]. Note that for a better clarity of presentation, the coordinate axes in the panels are not to the same scale. See the online version for plots in colour.

6. Concluding remarks

The central idea of this work was to identify the best generalised invariant \mathcal{J}_α so that a material can easily be described by a strain-energy function of a single argument $W = W(\mathcal{J}_\alpha)$. Remarkably, our generalised invariant supports pseudo-universal relationships. In turn, these relationships can be used to find the best possible value of α without prescribing the functional form of W .

The main practical implications of these new *pseudo-universal* relationships, showcased in Section 4, are: (i) the application of the pseudo-universal relationships to experimental data will help *a priori* fixing the exponent of the principal stretches, and thereby identifying the appropriate class of models that may be suitable for modelling the deformation of the given specimen/dataset; and (ii) the constitutive parameter α can be quantified *a priori* before choosing a model W . Hence, our method has two main advantages: (i) it will reduce the likelihood of the non-uniqueness of the obtained fits and the identified model parameter

values in the process of fitting; and (ii) it will streamline the optimisation process, as at least one constitutive parameter (α) does not require to be included in the optimisation process.

Another important observation is the remarkable fact that values of the exponent α obtained from different deformation datasets of a specimen are close to each other, suggesting that the exponent α captures a generic intrinsic feature of the microstructure. To further investigate this finding, we should extend this modelling exercise to datasets with multiaxial deformations (i.e., not just the uniaxial deformation) and use pre-determined values of α obtained from the pseudo-universal relationships.

On identifying the value of the exponent α , and thereby informing the choice of a suitable strain energy function, it is likely (as was the case for the exemplar specimens/datasets in Section 5) that the appropriate class of models to be considered for the finite deformation of soft materials may be that of the form $W(\mathcal{J})$. The results in Section 5.2 indicate that in this class of models the choice of the functional form of the model still remains an important step. The seminal Ogden model, as an archetypical example of models in this class, appeared to run into difficulty in capturing the deformation behaviour of the considered specimens with a single monomial invariant \mathcal{J} , whereas the parent model given by Eq. (19) captured the datasets favourably with the same monomial invariant \mathcal{J} . This model is based on a higher-order rational approximation than most existing models (see Anssari-Benam, 2021). As pointed out by Destrade et al. (2017), constructing strain-energy functions based on higher order rational approximants may open up the possibility of a new class of models whose functional forms may accommodate a more generalised form of the invariant \mathcal{J} .

Our approach also raises the possibility of looking at a large set of different strain-energy functions using automatic model discovery through machine learning as recently proposed by Linka and Kuhl (2023), Linka et al. (2023a; 2023b), Martonová et al. (2024) and Peirlinck et al. (2024). Such an investigation is likely to help bring this new method to the systematic study of soft tissues and other practical applications.

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Declaration of Competing Interest The authors have no competing interests to declare.

Data Availability The numerical values of the experimental datapoints used in this work have been provided in Tables A1 and A2 of Appendix A, and in Kawabata et al. (1981), as indicated in the manuscript.

Appendix A Tabulated numerical datapoints of the datasets used in this work

Table A1 – Numerical datapoints for the uniaxial and pure shear deformations of the rubber specimens from Jones and Treloar (1975).

Uniaxial deformation		Pure shear deformation		
λ_{uni} [-]	T_{uni} [MPa]	λ_{ps} [-]	$(T_{ps})_1$ [MPa]	$(T_{ps})_2$ [MPa]
1	0	1	0	0
1.02	0.04	1.05	0.10 ₅	0.04
1.04	0.07	1.10	0.21	0.08
1.08	0.14	1.15	0.31	0.12
1.13	0.20	1.20	0.42	0.16
1.17 ₅	0.25	1.30	0.66	0.21
1.31	0.44	1.40	0.92	0.25
1.53	0.75 ₅	1.50	1.21 ₅	0.29
1.71	1.00	1.60	1.53	0.33
1.96	1.38	1.70	1.88	0.36
2.20 ₅	1.75	1.80	2.26	0.39
2.35	2.03	1.90	2.68	0.41
2.47	2.28	2.00	3.14	0.43
2.61 ₅	2.55	2.10	3.65	0.45 ₅
		2.20	4.21 ₅	0.47 ₅
		2.30	4.82 ₅	0.49
		2.40	5.48	0.51
		2.50	6.18	0.53
		2.60	6.93	0.55
		2.70	7.72 ₅	0.56
		2.80	8.56	0.58
		2.85	9.01	0.59

Table A2 – Numerical datapoints for the uniaxial and pure shear deformations of carbon black reinforced natural rubber vulcanizates due to Fukahori and Seki (1992).

Uniaxial deformation		Pure shear deformation		
λ_{uni} [-]	T_{uni} [MPa]	λ_{ps} [-]	$(T_{ps})_1$ [MPa]	$(T_{ps})_2$ [MPa]
0.30	-1.49	1	0	0
0.40 ₅	-1.24 ₅	1.25 ₅	0.53	0.19
0.49	-0.87	1.48	0.97	0.31
0.58 ₅	-0.67	1.74 ₅	1.44	0.38 ₅
0.69	-0.47	2.00	2.00	0.44
0.80	-0.31 ₅	2.27 ₅	2.71	0.48
0.91	-0.13	2.53	3.55	0.54
1.00	0	2.80	4.64	0.58
1.23	0.40	3.06	6.00	0.61 ₅
1.50 ₅	0.81	3.31	7.77 ₅	0.71
1.77 ₅	1.27	3.57	9.88	0.79
2.04 ₅	1.90	4.10	16.00	1.04
2.58 ₅	3.42	4.63	24.56	1.36 ₅
3.10	5.87	5.00	32.88 ₅	1.67
3.65	9.64			
4.19	15.41			
4.73	23.75			
5.00	29.28 ₅			

Table A3 – Numerical datapoints for the biaxial deformation of rubber specimens due to Jones and Treloar (1975), for four biaxial deformation paths.

$\lambda_2 = 1.502$			$\lambda_2 = 1.984$			$\lambda_2 = 2.295$			$\lambda_2 = 2.623$		
λ_1 [-]	T_1 [MPa]	T_2 [MPa]	λ_1 [-]	T_1 [MPa]	T_2 [MPa]	λ_1 [-]	T_1 [MPa]	T_2 [MPa]	λ_1 [-]	T_1 [MPa]	T_2 [MPa]
0.82	0	1.06	0.71	0	2.82	0.66	0	4.45	0.62	0	6.79
0.85	0.05	1.10	0.75	0.05	2.88	0.70	0.05	4.50	0.65	0.03 ₅	6.85
0.90	0.13	1.15	0.80	0.12	2.94	0.75	0.11	4.56	0.70	0.09 ₅	6.93
0.95	0.21 ₅	1.20	0.85	0.19	3.00	0.80	0.18	4.61	0.75	0.16	7.01 ₅
1.00	0.31	1.23 ₅	0.90	0.27	3.04 ₅	0.85	0.25	4.66 ₅	0.80	0.23	7.09
1.10	0.50	1.30 ₅	0.95	0.36	3.09	0.90	0.33	4.71	0.85	0.31	7.16
1.20	0.71 ₅	1.36 ₅	1.00	0.45	3.13	0.95	0.42	4.76	0.90	0.39	7.22
1.30	0.96	1.41	1.10	0.65	3.21	1.00	0.51	4.81	0.95	0.48	7.28
1.40	1.23 ₅	1.457	1.20	0.87	3.28	1.10	0.71	4.90	1.00	0.57	7.33
1.50	1.54	1.50	1.30	1.12	3.33 ₅	1.20	0.94	4.98 ₅	1.10	0.77	7.43
1.60	1.86	1.53	1.40	1.39	3.39	1.30	1.18 ₅	5.06	1.20	1.00	7.52
1.70	2.22	1.57	1.50	1.70	3.44	1.40	1.51	5.15	1.30	1.27	7.61
1.80	2.62	1.60	1.60	2.04	3.49	1.50	1.77	5.23	1.40	1.57	7.70
1.90	3.05	1.63	1.70	2.42	3.54	1.60	2.12	5.30 ₅	1.50	1.90	7.79
2.00	3.53	1.66	1.80	2.84	3.58	1.70	2.51	5.38	1.60	2.26	7.89
2.10	4.05	1.69	1.90	3.31	3.63	1.80	2.95	5.46	1.70	2.66	7.99
2.20	4.61	1.72	2.00	3.81	3.69	1.90	3.42	5.53	1.80	3.11	8.10
2.30	5.30	1.75 ₅	2.10	4.36	3.75	2.00	3.93	5.61	1.90	3.61	8.20
2.40	5.97	1.79	2.20	4.97	3.81	2.10	4.48	5.69	2.00	4.17	8.32
2.50	6.73	1.82	2.30	5.66	3.89	2.20	5.08	5.76	2.10	4.77	8.43
2.60	7.55	1.85				2.30	5.72 ₅	5.84	2.20	5.43	8.55
2.62 ₅	7.76 ₅	1.86							2.30	6.14	8.67
									2.40	6.92	8.79
									2.50	7.81	8.92
									2.60	8.78	9.05
									2.62 ₅	9.05 ₅	9.08

References

- Anssari-Benam, A., 2021. On a new class of non-Gaussian molecular-based constitutive models with limiting chain extensibility for incompressible rubber-like materials. *Math. Mech. Solids* 26, 1660–1674. <https://doi.org/10.1177/10812865211001094>.
- Anssari-Benam, A., 2023. Large isotropic elastic deformations: On a *comprehensive* model to correlate the theory and experiments for incompressible rubber-like materials. *J. Elast.* 153, 219–244. <https://doi.org/10.1007/s10659-022-09982-5>.
- Anssari-Benam, A., Bucci, A., Saccomandi, G., 2021. On the central role of the invariant I_2 in nonlinear elasticity. *Int. J. Eng. Sci.* 163, 103486. <https://doi.org/10.1016/j.ijengsci.2021.103486>.
- Beatty, M.F., 2003. An average-stretch full-network model for rubber elasticity. *J. Elast.* 70, 65–86. <https://doi.org/10.1023/B:ELAS.0000005553.38563.91>.
- Criscione, J.C., Humphrey, J.D., Douglas, A.S., Hunter, W.C., 2000. An invariant basis for natural strain which yields orthogonal stress response terms in isotropic hyperelasticity. *J. Mech. Phys. Solids* 48, 2445–2465. [https://doi.org/10.1016/S0022-5096\(00\)00023-5](https://doi.org/10.1016/S0022-5096(00)00023-5).
- Destrade, M., Gilchrist, M.D., Murphy, J.G., Rashid, B., Saccomandi, G., 2015. Extreme softness of brain matter in simple shear. *Int. J. Non-Linear Mech.* 75, 54–58. <https://doi.org/10.1016/j.ijnonlinmec.2015.02.014>.
- Destrade, M., Saccomandi, G., Sgura, I., 2017. Methodical fitting for mathematical models of rubber-like materials. *Proc. R. Soc. A* 473, 20160811. <https://doi.org/10.1098/rspa.2016.0811>.
- Ennis, D.B., Kindlmann, G., 2006. Orthogonal tensor invariants and the analysis of diffusion tensor magnetic resonance images. *Magn. Reson. Med.* 55, 136–146. <https://doi.org/10.1002/mrm.20741>.
- Fried, E., 2002. An elementary molecular-statistical basis for the Mooney and Rivlin–Saunders theories of rubber elasticity. *J. Mech. Phys. Solids* 50, 571–582. [https://doi.org/10.1016/S0022-5096\(01\)00086-2](https://doi.org/10.1016/S0022-5096(01)00086-2).
- Fukahori, Y., Seki, W., 1992. Molecular behaviour of elastomeric materials under large deformation: 1. Re-evaluation of the Mooney–Rivlin plot. *Polymer* 33, 502–508. [https://doi.org/10.1016/0032-3861\(92\)90726-D](https://doi.org/10.1016/0032-3861(92)90726-D).
- Gent, A.N., 1996. A new constitutive relation for rubber. *Rubber Chem. Technol.* 69, 59–61. <https://doi.org/10.5254/1.3538357>.
- Horgan, C.O., Murphy, J.G., 2023. Pure torsion for incompressible hyperelastic materials of Valanis–Landel form. *Proc. R. Soc. A* 479, 20230011. <https://doi.org/10.1098/rspa.2023.0011>.
- Horgan, C.O., Saccomandi, G., 1999. Simple torsion of isotropic, hyperelastic, incompressible materials with limiting chain extensibility. *J. Elast.* 56, 159–170. <https://doi.org/10.1023/A:1007606909163>.
- Horgan, C.O., Smayda, M.G., 2012. The importance of the second strain invariant in the constitutive modeling of elastomers and soft biomaterials. *Mech. Mater.* 51, 43–52. <https://doi.org/10.1016/j.mechmat.2012.03.007>.
- Jones, D.F., Treloar, L.R.G., 1975. The properties of rubber in pure homogeneous strain. *J. Phys. D: Appl. Phys.* 8, 1285–1304. <https://doi.org/10.1088/0022-3727/8/11/007>.
- Kawabata, S., Matsuda, M., Tei, K., Kawai, H., 1981. Experimental survey of the strain energy density function of isoprene rubber vulcanizate. *Macromolecules* 14, 154–162. <https://doi.org/10.1021/ma50002a032>.
- Kearsley, E.A., 1989. Note: Strain invariants expressed as average stretches. *J. Rheol.* 33, 757–760. <https://doi.org/10.1122/1.550063>.
- Khiêm, V.N., Itskov, M., 2016. Analytical network-averaging of the tube model: Rubber elasticity. *J. Mech. Phys. Solids* 96, 254–269. <https://doi.org/10.1016/j.jmps.2016.05.030>.
- Kuhl, E., Goriely, A., 2024. I too ♥ I_2 : A new class of hyperelastic isotropic incompressible models based solely on the second invariant. *J. Mech. Phys. Solids* 188, 105670. <https://doi.org/10.1016/j.jmps.2024.105670>.
- Linka, K., Buganza Tepole, A., Holzapfel, G.A., Kuhl, E., 2023a. Automated model discovery for skin: Discovering the best model, data, and experiment. *Comput. Methods Appl. Mech. Eng.* 410, 116007. <https://doi.org/10.1016/j.cma.2023.116007>.
- Linka, K., Kuhl, E., 2023. A new family of Constitutive Artificial Neural Networks towards automated model discovery. *Comput. Methods Appl. Mech. Eng.* 403, 115731. <https://doi.org/10.1016/j.cma.2022.115731>.
- Linka, K., St. Pierre, S.R., Kuhl, E., 2023b. Automated model discovery for human brain using Constitutive Artificial Neural Networks. *Acta Biomater.* 160, 134–151. <https://doi.org/10.1016/j.actbio.2023.01.055>.
- Martonová, D., Peirlinck, M., Linka, K., Holzapfel, G.A., Leyendecker, S., Kuhl, E., 2024. Automated model discovery for human cardiac tissue: Discovering the best model and parameters. *Comput. Methods Appl. Mech. Eng.* 428, 117078. <https://doi.org/10.1016/j.cma.2024.117078>.
- Mihai, L.A., Goriely, A., 2017. How to characterize a nonlinear elastic material? A review on nonlinear constitutive parameters in isotropic finite elasticity. *Proc. R. Soc. A* 473, 20170607. <https://doi.org/10.1098/rspa.2017.0607>.
- Murphy, J.G., 2006. Some remarks on kinematic modeling of limiting chain extensibility. *Math. Mech. Solids* 11, 629–641. <https://doi.org/10.1177/10812865050523>.
- Ogden, R.W., 1972. Large deformation isotropic elasticity – on the correlation of theory and experiment for incompressible rubberlike solids. *Proc. R. Soc. Lond. A* 326, 565–584. <https://doi.org/10.1098/rspa.1972.0026>.
- Ogden, R.W., Saccomandi, G., Sgura, I., 2004. Fitting hyperelastic models to experimental data. *Comput. Mech.* 34, 484–502. <https://doi.org/10.1007/s00466-004-0593-y>.
- Peirlinck, M., Linka, K., Hurtado, J.A., Kuhl, E., 2024. On automated model discovery and a universal material subroutine for hyperelastic materials. *Comput. Methods Appl. Mech. Eng.* 418, 116534. <https://doi.org/10.1016/j.cma.2023.116534>.
- Pucci, E., Saccomandi, G., 1997. On universal relations in continuum mechanics. *Continuum Mech. Thermodyn.* 9, 61–72. <https://doi.org/10.1007/s001610050055>.
- Rivlin, R.S., 1948. Large elastic deformations of isotropic materials IV. further developments of the general theory. *Philos. Trans. R. Soc. Lond. A* 241, 379–397. <https://doi.org/10.1098/rsta.1948.0024>.

- Rivlin, R.S., Sawyers, K.N., 1976. The strain-energy function for elastomers. *Trans. Soc. Rheol.* 20, 545–557. <https://doi.org/10.1122/1.549436>.
- Saccomandi, G., 2001. Universal results in finite elasticity, in: Fu, Y., Ogden, R.W. (Eds.), *Nonlinear Elasticity: Theory and Applications*. Cambridge University Press, Cambridge, UK, p. 97–134. <https://doi.org/10.1017/CB09780511526466.004>.
- Saccomandi, G., Vergori, L., 2019. Generalised Mooney–Rivlin models for brain tissue: A theoretical perspective. *Int. J. Non-Linear Mech.* 109, 9–14. <https://doi.org/10.1016/j.ijnonlinmec.2018.09.008>.
- Saccomandi, G., Vergori, L., 2021. Some remarks on the weakly nonlinear theory of isotropic elasticity. *J. Elast.* 147, 33–58. <https://doi.org/10.1007/s10659-021-09865-1>.
- Valanis, K.C., Landel, R.F., 1967. The strain-energy function of a hyperelastic material in terms of the extension ratios. *J. Appl. Phys.* 38, 2997–3002. <https://doi.org/10.1063/1.1710039>.
- Wineman, A., 2005. Some results for generalized neo-Hookean elastic materials. *Int. J. Non-Linear Mech.* 40, 271–279. <https://doi.org/10.1016/j.ijnonlinmec.2004.05.007>.