Comparison of hyperelastic models for rubber-like materials

Gilles Marckmann, Erwan Verron

COMPARISON OF HYPERELASTIC MODELS FOR RUBBERLIKE MATERIALS

G. Marckmann and E. Verron
Institut de Recherche en Génie Civil et Mécanique,
UMR CNRS 6183, École Centrale de Nantes,
BP 92101, 44321 Nantes cedex 3, France.

ABSTRACT

The present paper proposes a thorough comparison of twenty hyperelastic models for rubberlike materials. The ability of these models to reproduce different types of loading conditions is analyzed thanks to two classical sets of experimental data. Both material parameters and the stretch range of validity of each model are determined by an efficient fitting procedure. Then, a ranking of these twenty models is established, highlighting new efficient constitutive equations that could advantageously replace well-known models, which are widely used by engineers for finite element simulation of rubber parts.

INTRODUCTION

Elastomeric materials are used in automotive parts such as tires, engine and transmission mounts, center bearing supports and exhaust rubber parts. Nowadays, the design of these highly technical parts necessitates the use of simulation tools such as finite element softwares. In this context, an appropriate constitutive model is an essential prerequisite for good numerical predictions. There was a significant number of papers which proposed new constitutive equations for rubber in the last few years. The general theory of non-linear hyperelasticity is classically invoked to predict the response of parts under static loading conditions [1] or to develop more sophisticated models for viscoelasticity or stress-softening (see for example [2–7]).

Many models have been proposed to describe the elastic response of elastomers, but only few of them are revealed able to describe the complete behavior of the material, i.e. to satisfactorily reproduce experimental data for different loading conditions (uniaxial or biaxial extension, simple or pure shear). In the following, the expression *complete behavior* refers to the response of the material under different loading types. Obviously, the most interesting models are those which can describe this complete behavior with the minimal number of material parameters which should be experimentally determined. Nevertheless, it is often difficult for an engineer to choose between existing models.

Few studies evaluate and compare the ability of hyperelastic models to reproduce the complete behavior of elastomers. Some authors demonstrate the efficiency of their own model especially for large strain [8,9], or compare one model to another in order to establish equivalence of formulations [10,11]. Recently, Seibert and Schöche [12] compared six different models considering

their own experimental data obtained with uniaxial and biaxial extension tests. The danger of series formulations is highlighted by poor predictions of the biaxial response of models after having determined the material parameters with uniaxial experimental data. Boyce and Arruda[13] compared five models using Treloar's experimental data[14] for three different types of deformation (uniaxial, biaxial and pure shear). More recently, Attard and Hunt considered experimental data of seven different authors for uniaxial tension, pure shear, equibiaxial tension, compression and biaxial extension to demonstrate the efficiency of their model[15].

The present paper proposes a thorough comparison of twenty hyperelastic models and a classification of them with respect to their ability to fit experimental data. After recalling basic notation, the formulation of each model considered here is briefly summarized. Then, experimental data and methods adopted to determine material parameters are described. Afterwards, comparison criteria and the corresponding ranking of models are established. Final remarks close the paper.

PRELIMINARY REMARK

Throughout the rest of the paper, elastomers are assumed isotropic and incompressible, and all inelastic phenomena such as viscoelasticity, stress-softening or damage are neglected. Only their highly non-linear elastic response under large strain is retained and the general theory of hyperelasticity is considered.

BASICS OF CONTINUUM MECHANICS

In the following, strain and stress tensors are first briefly recalled. Then, the general formulation of non-linear incompressible hyperelasticity is derived. For details, the reader can refer for example to[16] and[17].

DEFORMATION TENSORS

Consider the deformation of a rubberlike solid and denote $\mathbf{F}$ the local gradient of the deformation. The right and left Cauchy-Green deformation tensors, respectively $\mathbf{C}$ and $\mathbf{B}$, are defined by:

$$\mathbf{C} = \mathbf{F}^t\,\mathbf{F} \quad \text{and} \quad \mathbf{B} = \mathbf{F}\,\mathbf{F}^t. \tag{1}$$

$\mathbf{C}$ and $\mathbf{B}$ admit the three same principal invariants classically denoted I_1, I_2 and I_3 and given by:

$$I_1 = \mathrm{tr}\,(\mathbf{C}) \tag{2}$$

$$I_2 = \frac{1}{2}\left[\mathrm{tr}\,(\mathbf{C})^2 - \mathrm{tr}\left(\mathbf{C}^2\right)\right] \tag{3}$$

$$I_3 = \det \mathbf{C}. \tag{4}$$

In these equations, $\mathbf{C}$ can be replaced by $\mathbf{B}$. Stretch ratios are defined as the square roots of the eigenvalues of $\mathbf{C}$ (equal to those of $\mathbf{B}$) and are classically

denoted $(\lambda_i)_{i=1,3}$. Using these ratios, principal invariants reduce to:

$$I_1 = \lambda_1^2 + \lambda_2^2 + \lambda_3^2 \quad (5)$$
$$I_2 = \lambda_1^2\lambda_2^2 + \lambda_2^2\lambda_3^2 + \lambda_3^2\lambda_1^2 \quad (6)$$
$$I_3 = \lambda_1^2\lambda_2^2\lambda_3^2. \quad (7)$$

STRESS TENSORS

Stresses are internal cohesion forces inside the matter. For large strain problems, two major stress tensors are classically defined: the true (or Cauchy) stress tensor $\boldsymbol{\sigma}$ and the nominal (or first Piola-Kirchhoff) stress tensor $\boldsymbol{P}$. They are related by:

$$\boldsymbol{P} = \det\mathbf{F}\,\boldsymbol{\sigma}\,\mathbf{F}^{-t}, \quad (8)$$

in which the exponent $\cdot^{-t}$ denotes the transposition of the inverse.

INCOMPRESSIBLE HYPERELASTIC CONSTITUTIVE EQUATIONS

In the general theory of hyperelasticity, it is assumed that stress tensors derive from strain energy function, which is defined per unit of undeformed volume, depends on the strain tensor $\mathbf{B}$ and is classically denoted W. Considering incompressible materials leads to a kinematical condition on strain:

$$I_3 = 1. \quad (9)$$

Consequently, stress tensors depend on both strain and an arbitrary scalar parameter p which can be determined with equilibrium equations:

$$\boldsymbol{\sigma} = 2\mathbf{B}\frac{\partial W}{\partial \mathbf{B}} - p\mathbf{I} \quad (10)$$

where $\mathbf{I}$ is the identity tensor, and:

$$\boldsymbol{P} = \frac{\partial W}{\partial \mathbf{F}} - p\mathbf{F}^{-t}. \quad (11)$$

Assuming now that the material is isotropic, the strain energy function only depends on the two first strain invariants and stress tensors can be written as[17]:

$$\boldsymbol{\sigma} = 2\left(\frac{\partial W}{\partial I_1} + I_1\frac{\partial W}{\partial I_2}\right)\mathbf{B} - 2\frac{\partial W}{\partial I_2}\mathbf{B}^2 - p\mathbf{I} \quad (12)$$

and

$$\mathbf{P} = 2\mathbf{F}\left(\left[\frac{\partial W}{\partial I_1} + I_1\frac{\partial W}{\partial I_2}\right]\mathbf{I} - \frac{\partial W}{\partial I_2}\mathbf{C}\right) - p\mathbf{F}^{-t}. \quad (13)$$

Finally, principal stress can be determined in terms of principal stretch ratios:

$$\sigma_i = 2\left(\lambda_i^2\frac{\partial W}{\partial I_1} - \frac{1}{\lambda_i^2}\frac{\partial W}{\partial I_2}\right) - p \qquad i = 1,3 \quad (14)$$

and:

$$P_i = 2\left(\lambda_i\frac{\partial W}{\partial I_1} - \frac{1}{\lambda_i^3}\frac{\partial W}{\partial I_2}\right) - p\frac{1}{\lambda_i} \qquad i = 1,3. \quad (15)$$

SIMPLE LOADING CONDITIONS

Using the previous Eq. (15), the stress-stretch relationships corresponding to simple tests can be easily derived:

- for uniaxial extension:

$$P = 2\left(\lambda - \frac{1}{\lambda^2}\right)\left(\frac{\partial W}{\partial I_1} + \frac{\partial W}{\partial I_2}\frac{1}{\lambda}\right), \tag{16}$$

- for equibiaxial extension:

$$P = 2\left(\lambda - \frac{1}{\lambda^5}\right)\left(\frac{\partial W}{\partial I_1} + \frac{\partial W}{\partial I_2}\lambda^2\right), \tag{17}$$

- for pure shear:

$$P = 2\left(\lambda - \frac{1}{\lambda^3}\right)\left(\frac{\partial W}{\partial I_1} + \frac{\partial W}{\partial I_2}\right) \tag{18}$$

- for biaxial extension:

$$P_1 = 2\left(\lambda_1 - \frac{1}{\lambda_1^3\lambda_2^2}\right)\left(\frac{\partial W}{\partial I_1} + \frac{\partial W}{\partial I_2}\lambda_2^2\right) \tag{19}$$

and

$$P_2 = 2\left(\lambda_2 - \frac{1}{\lambda_1^2\lambda_2^3}\right)\left(\frac{\partial W}{\partial I_1} + \frac{\partial W}{\partial I_2}\lambda_1^2\right). \tag{20}$$

In these equations, P and λ represent the nominal stress and the stretch measured during the experiments. In the case of biaxial extension, P_1 and P_2 (respectively λ_1 and λ_2) stand for the nominal in-plane stress (resp. in-plane stretches). In every case, the plane stress condition is adopted such as $P_3 = 0$.

CONSTITUTIVE MODELS

Hyperelastic models are classified into three types of formulation, depending on the approach followed by the authors to develop the strain energy function:

- the first kind of models are issued from mathematical developments of W such as the well-known Rivlin series[18] or the Ogden real exponents[19]. They are classically referred as phenomenological models. Material parameters are generally difficult to determine and such models can lead to error when they are used out of the deformation range in which their parameters were identified,
- other authors, such as Rivlin and Saunders[20], and Hart-Smith[21], directly determine the material functions $\partial W/\partial I_1$ and $\partial W/\partial I_2$ using experimental data,
- the third kind of models are those developed from physical motivation. Such models are based on both physics of polymer chains network and statistical methods. It leads to different strain energy functions depending on microscopic phenomena accounted for. In most of the cases, their mathematical formulation is quite complicated.

The Mooney model. Mooney [22] observed that rubber response is linear under simple shear loading conditions. He considers W under the following form:

$$W = C_1(I_1 - 3) + C_2(I_2 - 3) \tag{21}$$

where C_1 and C_2 are the two material parameters. This model is widely used for rubber parts in which deformation remains moderate (lower than 200%).

The Mooney-Rivlin model. Rivlin [18,23] extended the previous model by developing W as a polynomial series of $(I_1 - 3)$ and $(I_2 - 3)$:

$$W = \sum_{i=0,j=0}^{\infty} C_{ij}(I_1 - 3)^i (I_2 - 3)^j \tag{22}$$

where C_{ij} are material parameters and $C_{00} = 0$. The series is often truncated to terms of the second or third order [24–26]. As an example, a third order truncation necessitates the determination of 9 material parameters. For some authors, the so-called Rivlin representation of W can be improved by considering other strain invariants [15,27]. Nevertheless, this form of strain energy is classically used for very large strain problems.

The Biderman model. In the previous series Eq. (22), Biderman [28] only retained terms for which $i = 0$ or $j = 0$; he considered the first three terms for I_1 and only one term for I_2:

$$W = C_{10}(I_1 - 3) + C_{01}(I_2 - 3) + C_{20}(I_1 - 3)^2 + C_{30}(I_1 - 3)^3. \tag{23}$$

This model was successfully used by Alexander [29].

The Haines-Wilson model. Comparing invariants and principal stretches developments of W, James *et al.* [25] chose to retain only six terms of the series:

$$\begin{aligned} W = {} & C_{10}(I_1 - 3) + C_{01}(I_2 - 3) + C_{11}(I_1 - 3)(I_2 - 3) \\ & + C_{02}(I_2 - 3)^2 + C_{20}(I_1 - 3)^2 + C_{30}(I_1 - 3)^3. \end{aligned} \tag{24}$$

The Ogden model. In 1972, Ogden [19] proposed to derive W in terms of generalized strain [30]. He expanded the strain energy through a series of real powers of $(\lambda_i)_{i=1,3}$:

$$W = \sum_{n=1}^{N} \frac{\mu_n}{\alpha_n} \left(\lambda_1^{\alpha_n} + \lambda_2^{\alpha_n} + \lambda_3^{\alpha_n} - 3\right) \tag{25}$$

where the material parameters $(\mu_n, \alpha_n)_{n=1,N}$ should fulfilled the following stability condition:

$$\mu_n \alpha_n > 0 \qquad \forall\ n = 1, N. \tag{26}$$

Considering experimental data of Treloar [14], the author proposed a 6 parameters model ($N = 3$) which leads to excellent agreement with simple tension, pure shear and equibiaxial tension data. This model is one of the most widely used for large strain problems, even if the determination of material parameters leads to some difficulties.

The Shariff model. Recently, Shariff[31] proposed a new model for which W takes the form of a function series. He considers a separable form of the strain energy function in terms of the principal stretch ratios:

$$\sigma_i = -p + \lambda_i \frac{\partial W}{\partial \lambda_i} = f(\lambda_i), \tag{27}$$

where f is a series of regular functions ϕ_j. Parameters α_j are linear coefficients of these functions and functions ϕ_j are chosen in order to satisfy the linear theory of incompressible isotropic elasticity for all values of α_j. For this reason, the Young modulus is proposed as a general factor and f can be written as:

$$f(\lambda) = E \sum_{j=0}^{n} \alpha_j \phi_j(\lambda) \tag{28}$$

with $\alpha_0 = 1$. Then, the author proposes the following values for ϕ_j:

$$\begin{aligned}
\phi_0(\lambda) &= \frac{2\ln(\lambda)}{3} \\
\phi_1(\lambda) &= e^{(1-\lambda)} + \lambda - 2 \\
\phi_2(\lambda) &= e^{(\lambda-1)} - \lambda \\
\phi_3(\lambda) &= \frac{(\lambda-1)^3}{\lambda^{3.6}} \\
\phi_j(\lambda) &= (\lambda-1)^{j-1}, \qquad j = 4, 5, ..., n.
\end{aligned} \tag{29}$$

In order to satisfy the polyconvexity of the strain energy function, Shariff adds stability conditions on the range of scalars α_j.

EXPERIMENTAL DETERMINATION OF $\partial W/\partial I_1$ AND $\partial W/\partial I_2$

The Rivlin and Saunders model. Rivlin and Saunders[20] used a biaxial tensile tester to obtain experimental conditions for which I_1 or I_2 are set constant. They observed that, for a carbon black filled natural rubber, $\partial W/\partial I_1$ does not depend on I_1 and I_2, and $\partial W/\partial I_2$ does not depend on I_1. They also showed that the ratio $\frac{\partial W}{\partial I_2}/\frac{\partial W}{\partial I_1}$ decreases with I_2 and they proposed to consider W under the following form:

$$W = C(I_1 - 3) + f(I_2 - 3) \tag{30}$$

where the function f has to determined thanks to experimental data.

The Gent and Thomas model. Considering the general form proposed by Rivlin and Saunders (Eq. (30)), Gent and Thomas[32] proposed the following empirical strain energy function which involves only two material parameters:

$$W = C_1(I_1 - 3) + C_2 \ln\left(\frac{I_2}{3}\right). \tag{31}$$

Nevertheless, this model is not revealed more efficient that the one proposed by Mooney (Eq. (21)).

The Hart-Smith model. Improving the results of Rivlin and Saunders, Hart-Smith[21] observed that $\partial W/\partial I_1$ is constant for values of I_1 smaller than 12, but that it increases for higher values of the first principal invariant. He explained this result by invoking the limit of extensibility of macromolecules which leads to the strain-hardening phenomenon observed during mechanical tests. Thus, he proposed to model this strain-hardening phenomenon using an exponential term in W:

$$\frac{\partial W}{\partial I_1} = G \exp\left[k_1 (I_1 - 3)^2\right] \quad \text{and} \quad \frac{\partial W}{\partial I_2} = G \frac{k_2}{I_2}. \tag{32}$$

The Valanis and Landel assumption. Valanis and Landel[33] suggested that an efficient function W had not been found before because of difficulties inherent in its dependence on strain invariants: functions $\partial W/\partial I_1$ and $\partial W/\partial I_2$ might be very complex and it is not easy to design experiments in which I_1 and I_2 are not interrelated. Then, they proposed to express W in terms of principal stretches $(\lambda_i)_{i=1,3}$ and they assume the strain separability of the strain energy function as:

$$W = w(\lambda_1) + w(\lambda_2) + w(\lambda_3). \tag{33}$$

Thus, the determination of W is restricted to the one of w. In the same paper, authors also proposed the following form of w (through the definition of its derivative):

$$\frac{dw}{d\lambda} = 2\mu \ln(\lambda) \tag{34}$$

The Gent model. Recently, Gent[34] invoked the concept of limiting chain extensibility to consider that I_1 should admit a maximum value denoted I_m, and he proposed the following strain energy function:

$$W = -\frac{E}{6}(I_m - 3) \ln\left[1 - \frac{I_1 - 3}{I_m - 3}\right] \tag{35}$$

where E and I_m are the two material parameters. Moreover, the author compared his approach with the physically-based model of Arruda and Boyce[35] (presented later in this paper).

The Yeoh and Fleming model. Yeoh[26] performed tensile, simple shear, compression and equibiaxial experiments and showed, like Rivlin and Saunders, and Hart-Smith before him, that $\partial W/\partial I_1$ is much greater than $\partial W/\partial I_2$. So, he proposed to neglect this second term. Later, Yeoh and Fleming[9] performed tensile tests on four different rubber materials. They observed that the reduced Mooney stress tends to a constant value which does not depend on I_1 for large strain ($I_1 \geq 8$). Consequently, they modified the Gent model (Eq. (35)) to propose a new strain energy function that involves three material parameters A, B and I_m:

$$W = \frac{A}{B}(I_m - 3)\left(1 - e^{-BR}\right) - C_{10}(I_m - 3)\ln(1 - R) \quad \text{with } R = \frac{(I_1 - 3)}{(I_m - 3)}. \tag{36}$$

PHYSICALLY-BASED MODELS

Physically-based models are founded on the microscopic response of polymer chains in the network. They differ one to each other depending on the assumptions made to reproduce this response.

The neo-Hookean model. The neo-Hookean model[36] is the simplest physically based constitutive equation for rubbers. It matches the Mooney-Rivlin model with only one material parameter ($C_2 = 0$ in Eq. (21)), but was derived from molecular chain statistics considerations. Rubber materials are constituted by a network of long flexible randomly oriented chains linked by chemical bounds at junction points[37]. The elasticity of this network is mainly due to entropic changes during deformation and the entropy of the material is defined by the number of possible conformations of macromolecular chains. In order to estimate the number of conformations, Treloar used a Gaussian statistical distribution and obtained the following form of W:

$$W = \frac{1}{2} nkT(I_1 - 3) \tag{37}$$

in which n is the chain density per unit of volume, k is the Boltzmann constant and T is the absolute temperature. For a carbon black-filled natural rubber, Treloar[14] obtained $\frac{1}{2}nkT = 0.2$ MPa. Then, his model was revealed in good agreement with tensile, simple shear and biaxial tests for deformation lower than 50%.

The 3-chain model. Before examining this model, let us briefly recall the concept of non-Gaussian chain elasticity. In 1942, Kuhn and Grün[38] used a non-Gaussian theory to take into account the limiting extensibility of polymer chains and they derived the strain energy of a single chain:

$$w = nkT \left[\frac{\lambda}{\sqrt{N}} \beta + \ln \frac{\beta}{\sinh \beta} \right] \quad \text{with} \quad \beta = \mathcal{L}^{-1} \left(\frac{\lambda}{\sqrt{N}} \right) \tag{38}$$

where $\mathcal{L}^{-1}$ denoted the inverse Langevin function define by $\mathcal{L}(x) = \coth(x) - 1/x$.

One year later, James and Guth[39] used the previous theory to derive a non-Gaussian constitutive equation for elastomers. They assumed that chains are randomly distributed and that the deformation of the network is driven by the gradient of the deformation (affine assumption). To simplify the transition between the strain energy of an individual chain and the one of the network, they proposed to consider that n chains are distributed upon the three principal strain axis with a density equal to $n/3$ in each direction. Thus, principal Cauchy stresses are given by:

$$\sigma_i = \frac{nkT}{3} \frac{\lambda}{\sqrt{N}} \, \lambda_i \, \mathcal{L}^{-1} \left(\frac{\lambda_i}{\sqrt{N}} \right) - p \tag{39}$$

Note that Flory[40] and later Treloar[41] developed similar models where the network chains are distributed upon four axis corresponding to directions of the vertices of a regular tetrahedron.

The Isihara model. Isihara[42] used the non-Gaussian theory and linearized the corresponding equations to obtain a Rivlin series form for W:

$$W = C_{10}(I_1 - 3) + C_{20}(I_1 - 3)^2 + C_{01}(I_2 - 3) \tag{40}$$

It can be noticed that this molecular model involves the second strain invariant I_2 which did not appear in earlier physically-based models. In this way, the Isihara model is close to the formulations of Biderman or Mooney-Rivlin.

The general theory of real chain network. The deviation in experimental data of the ideal chain models presented above is classically imputed to the so-called phantom assumption which does not account for chains entanglement and for which chains can pass through mutually. Authors like Flory, Ermann, Mark and Edwards among others[40,43–45] introduced the idea of entanglement constraints or topology conservation constraints. They proposed to separate the strain energy function as:

$$W = W_{ph} + W_c \tag{41}$$

where W_{ph} is the phantom network part and W_c is the constrained or cross-linking part. The three following models are based on this general theory.

The slip-link model. Ball *et al.*[45] developed the slip-link model by considering that chains are allowed to slip on a length a around a link. This model is mathematically complex:

$$W = \frac{1}{2}kTN_c\sum_{i=1}^{3}\lambda_i^2 + \frac{1}{2}kTN_s\sum_{i=1}^{3}\left[\frac{(1+\eta)\lambda_i^2}{1+\eta\lambda_i^2} + \ln|1+\eta\lambda_i^2|\right] \tag{42}$$

where N_c, N_s and η are the material parameters. We note that the first term of Eq. (42) corresponds to the phantom Gaussian model.

The van der Waals model. Kilian *et al.*[8,46] revived the idea of Wang and Guth by taking into account the van der Waals forces. The rubber network is treated as a gaz where interaction forces are applied between quasi-particules. The authors obtained the response of the material for different modes of deformation. Nevertheless, stress did not derive from a strain energy function. A few years later, the model is written in terms of strain energy by introducing a generalized invariant $\widetilde{I}$[47,48]:

$$W = G\left\{-(\lambda_m^2 - 3)\Big[\ln(1-\Theta) + \Theta\Big] - \frac{2}{3}\left(\frac{\widetilde{I}-3}{2}\right)^{\frac{3}{2}}\right\} \tag{43}$$

where $\Theta = \sqrt{(\widetilde{I}-3)/(\lambda_m^2-3)}$ and $\widetilde{I} = \beta I_1 + (1-\beta)I_2$. However, the material parameter β has no physical meaning, which confers to this model an empirical nature even if it is primarily based on molecular considerations.

The constrained junctions model. Flory and co-workers[40,43,49,50] developed a model based on Eq. (41) where junction points between chains are constrained to move in a restricted neighborhood due to other chains. The phantom part of the model is described by the neo-Hookean strain energy and the cross-linking part W_c is given by:

$$W_c = \frac{1}{2} kT\mu \sum_{i=1}^{3} \Big[B_i + D_i - \ln(B_i + 1) - \ln(D_i + 1) \Big] \tag{44}$$

with $B_i = \kappa^2(\lambda_i^2 - 1)(\lambda_i^2 + \kappa)^{-2}$ and $D_i = \lambda_i^2 \kappa^{-1} B_i$. This additional term improves the neo-Hookean model by leading better agreement with experimental data at moderate strain but this improvement is similar to the slip-link model. The use of the neo-Hookean model for the phantom part limits the constrained junctions model to stretches lower than 300% for uniaxial extension.

The 8-chain model. In 1993, Arruda and Boyce[35] proposed a chain model with a distribution of chains upon eight directions corresponding to the vertices of a cube inscribed in the unit sphere. This model is governed by the stretch of the diagonal of the cube $\lambda_{ch} = \sqrt{I_1/3}$. This simple model is isotropic and the principal Cauchy stresses are:

$$\sigma_i = \frac{nkT\sqrt{N}}{3} \, \frac{\lambda_i^2}{\lambda_{ch}} \, \mathcal{L}^{-1}\left(\frac{\lambda_{ch}}{\sqrt{N}} \right). \tag{45}$$

The product nkT is the first material parameter and is generally noted C_r. This model is quite similar to the 3-chains model but presents better agreement with experimental data for equibiaxial extension.

The tube model. Heinrich and Kaliske[51] pursued the works of Edwards and Vilgis[52], and Doi[53]. They proposed a model in which chains are constrained to remain in a tube formed by surrounding chains. This assumption is attributed to the high degree of entanglement of the rubber network. The confinement of chains is governed by a topology restoring potential. The authors used the statistical mechanics to determine this potential:

$$W = G_c \, I^*(2) - \frac{2G_e}{\beta} I^*(-\beta) \tag{46}$$

where $I^*(\alpha)$ is the first invariant of the generalized α-order strain tensor. The model takes the form of the two terms Ogden model with $\alpha_1 = 2$, $\alpha_2 = -\beta$, $\mu_1 = G_c$ and $\mu_2 = -2G_e/\beta$. However, this model is limited to moderate deformation and is not able to reproduce strain-hardening.

The extended-tube model. Limitations of the above model to moderate deformations are inherent to its foundations which refer to entanglement constraints but not to chain extensibility. Kaliske and Heinrich[54] replaced the Gaussian distribution by the non-Gaussian one, they introduced an inextensibility parameter δ and established a new strain energy function in which

the cross-link part is:

$$W_c = \frac{G_c}{2}\left[\frac{(1-\delta^2)(I_1-3)}{1-\delta^2(I_1-3)} + \ln(1-\delta^2(I_1-3))\right] \tag{47}$$

while the tube constraint term of Eq. (46) remains unchanged. In the previous equation, the empirical parameter β is supposed to lie between 0 and 1.

The non-affine micro-sphere model. Very recently, Miehe *et al.*[55] developed an original approach by associating the full network model of Treloar[56] and Wu and Van der Giessen[57] with the tube-model of Heinrich *et al.*[51]. The numerical integration of individual chain contributions into the network is based on the work of Bazant[58]. The chains are continuously distributed in the unit sphere $\mathcal{S}$, and the integration over the surface of $\mathcal{S}$ is replaced by a discrete sum over m directions, denoted ${\mathbf{r}^i}_{i=1,m}$, with m corresponding weight factors ${w^i}_{i=1,m}$:

$$\int_{\mathcal{S}} v(A)p(A)dA \approx \sum_{i=1}^{m} v^i w^i \tag{48}$$

where v is the function to be integrated and $v^i = v(\mathbf{r}^i)$, $p(A)$ is the probability density function (constant if the distribution is uniform). The authors considered the Langevin free energy developed by Kuhn and Grün[38] for the chain response (Eq. (38)).

A non-affine model is proposed by allowing micro-stretches to fluctuate around macro-stretches. To this end, the p-root average of the non-affine stretch λ of the single polymer chain is set equal to the p-root average of the macroscopic stretch $\overline{\lambda}$, where p is an intrinsic parameter of the network. The corresponding model developed by Miehe *et al.* can be written with a phantom part W_{ph} and a tube contribution part W_c. In the stretch principal axis $\mathbf{e}^i$, the stress σ^i_{ph} of the phantom part contribution is expressed thanks to three material parameters μ, N, p:

$$\sigma^i_{ph} = \mu\sqrt{N}\lambda_i^2\lambda^{1-p}\left(\frac{\lambda}{\sqrt{N}}\right)\sum_{s=1}^{m} w^s\lambda_s^{p-2}(r_i^s)^2 \tag{49}$$

with

$$\lambda = \left[\sum_{s=1}^{m} w^s\lambda_s^{p/2}\right]^{1/p} \tag{50}$$

where $\lambda^s = \|\mathbf{F}\mathbf{r}^s\|$, $\mathbf{F}$ being the deformation gradient. The stress σ^i_c of the tube contribution also depends on three material parameters μ, U and q:

$$\sigma^i_c = -q\mu U\frac{1}{\lambda_i^2}\sum_{s=1}^{m} w^s(\overline{\nu}_s)^{q-2}(r_i^s)^2 \tag{51}$$

with $\overline{\nu}_s = \|{\mathbf{r}^s}^T\mathbf{C}^{-1}\mathbf{r}^s\|$. In Eqs (49-51), λ_i is the principal stretch in direction $\mathbf{e}^i$ and r_i^s is the i-th component of the s-th orientation vector $\mathbf{r}^s$. Considering incompressible materials, the hydrostatic pressure must be added to these terms. Authors suggested that a discretization of 21 directions on the half of the sphere is sufficient. They noted that for $p = 2$ and $q = 0$, the model reduices to the eight-chain model of Arruda and Boyce.

TABLE I: — LIST OF THE TWENTY MODELS COMPARED IN THE PRESENT PAPER SORTED BY THE YEAR OF PUBLICATION (N.M.P. STANDS FOR THE NUMBER OF MATERIAL PARAMETERS)

Model	Year	N.m.p	Parameters	Eqs
Mooney	1940	2	C_1, C_2	(21)
neo-Hookean	1943	1	$nkT/2$	(37)
3-chain	1943	2	$nkT/2$, N	(38)
Ishihara	1951	3	C_{10}, C_{01}, C_{20}	(40)
Biderman	1958	4	C_{10}, C_{01}, C_{20}, C_{30}	(23)
Gent and Thomas	1958	2	C_1, C_2	(31)
Hart-Smith	1966	3	G, k_1, k_2	(32)
Valanis and Landel	1967	1	μ	(34)
Ogden	1972	6	$(\mu_i, \alpha_i)_{i=1,3}$	(25)
Haines-Wilson	1975	6	C_{10}, C_{01}, ..., C_{30}	(24)
slip-link	1981	3	$N_c kT$, $N_S kT$, η	(42)
constrained junctions	1982	3	C_{10}, $kT\mu/2$, k	(44)
van der Waals	1986	4	G, a, λ_m, β	(43)
8-chain	1993	2	C_r, N	(45)
Gent	1996	2	E, I_m	(35)
Yeoh and Fleming	1997	4	A, B, C_{10}, I_m	(36)
tube	1997	3	G_c, G_e, β	(46)
extended-tube	1999	4	G_c, G_e, β, δ	(47)
Shariff	2000	5	E, $(\alpha_j)_{j=1,4}$	(28)
micro-sphere	2004	5	μ, N, p, U, q	(49), (51)

SUMMARY

The models which will be compared in the following are summarized in Table I.

DETERMINATION OF MATERIAL PARAMETERS

As mentioned in the introduction, it is now well-established that a unique experiment is not sufficient to characterize a rubber-like material even assuming that it is elastic[59–61]. Even if the fitting procedure converges for a given mechanical test, it is not ensured that other loading conditions will be well-reproduced with the same set of parameters. A good example is given in the paper of Seibert and Schöche[12].

With the incompressibility assumption, the admissible kinematical field of rubber-like materials is constrained. In the principal axes, this constraint leads that all deformation conditions are only governed by two independent variables, i.e. two independent stretch ratios. Then, relationships between equibiaxial extension and compression, and also pure and simple shear have already been established[14,62,63]. Therefore, a series of biaxial tests is revealed sufficient to completely characterize hyperelastic constitutive models.

EXPERIMENTAL DATA

In order to compare the efficiency of models, we choose two complementary data sets issued from classical references. The first set is due to Treloar[14]. It was widely used by other authors[13,15,19,21,29,35,64–66]. In the current study, data from Treloar[14] for unfilled natural rubber (cross-linked with 8 parts of S phr) was used. It exhibits highly reversible elastic response and no stretch-induced crystallization up to 400%. Thus it is well-modelled by hyperelastic constitutive equations. Experimental measures were performed for four different loading conditions: equibiaxial extension of a sheet (denoted EQE in the following), uniaxial tensile extension (denoted UE), pure shear (PS) and biaxial extension (denoted BE).

The second data set is due to Kawabata *et al.*[59]; it was obtained using an experimental apparatus for general biaxial extension testing. In terms of stretch ratios, unfilled polyisoprene specimen were stretched from 1.04 to 3.7 in the first direction (λ_1) and from 0.52 to 3.1 in the perpendicular direction (λ_2). These values correspond to moderate strain but lead to deformation conditions from uniaxial extension to equibiaxial extension.

Here, both data sets are simultaneously considered to compare models because the two materials are quite similar. Thus, for a given model, a unique set of material parameters must be able to reproduce these data with good agreement.

ALGORITHMS

The problem of determining material parameters consists in fitting theoretical solutions $\widehat{\mathbf{Y}}$ with experimental measures $\mathbf{Y}$. Experimental data are constituted of n points Y_i corresponding to n theoretical values $\widehat{Y}_i$. The discrepancy between theoretical and experimental results is classically defined in terms of the least square error given by:

$$\phi = \sum_{i=1}^{n} \|Y_i - \widehat{Y}_i\|^2 \tag{52}$$

In the above equation, weighting factors are sometimes added to moderate the influence of some particular data. So, if $\phi = 0$, experimental and theoretical values coincide. Nevertheless, as experimental data always exhibit some uncertainty and theoretical models depend on diverse assumptions, algorithms are always devoted to the minimization of ϕ instead of its annulment. In most of the cases, a residual discrepancy persists and the coincidence of $\widehat{\mathbf{Y}}$ with $\mathbf{Y}$ can only be established on a restrictive set of data. In the present case, this restriction leads to the reduction of the domain of the validity (in terms of stretching level) for the models.

Among all possible minimization algorithms, two different approaches are considered in the present study: classical gradient methods and genetic algorithms[67,68]. It is to note that the later has been used to determine material parameters only for few years[69–71]. More precisely, for a given model, material parameters are first determined using genetic algorithms; then, material parameters obtained with this method are used as initial guess of the classical Levenberg-Marquardt method[72,73]. If it does not converge then the mean square method is employed and if this latest approach also diverges then a

gradient method with variable step is used[74]. For more details on the use of these algorithms in the context of fitting constitutive models, the reader can refer to[63].

FITTING PROCEDURE

As the materials used by Treloar and Kawabata *et.al* are quite similar in terms of both composition and mechanical response, the aim of the fitting procedure is to determine if, for each model, a unique set of parameters is able to reproduce simultaneously the two sets of experimental data. Two fitting steps are performed to achieve this objective. For each model:

1. Parameters are determined with Treloar data for uniaxial tensile extension (UE), pure shear (PS), equibiaxial extension (EQE) and biaxial extension (BE).
   - 1.a. If the accuracy is good, parameters are retained.
   - 1.b. If the accuracy is poor, the domain of validity is reduced according to the following rules:
     - if the model is not able to reproduce strain-hardening observed for large strain, the domain of validity is reduced for uniaxial extension ($\lambda_{\max}$) and new parameters are determined for this new domain of validity,
     - elsewhere, data corresponding to other loading conditions (PS, EQE, BE) are progressively eliminated from the least-square error function by reducing their weighting factor in order to improve accuracy for uniaxial extension. Then, the domain of validity, i.e. $\lambda_{\max}$, for PS, EQE and BE is given on the response curves.
2. Parameters determined in the previous step are used to simulate Kawabata *et al.* biaxial experiments.
   - 2.a. If the accuracy is good, the parameters are considered as the appropriate parameters for both data sets.
   - 2.b. If not, new parameters are determined for the Kawabata *et al.* data using the Treloar parameters as initial guess for the procedure:
     - if the accuracy is not good, the domain of validity for biaxial extension is reduced,
     - elsewhere, the new parameters are retained for biaxial loading conditions and the domain of validity, i.e. λ_1 and λ_2, is given on the response curves.

RESULTS AND DISCUSSION

The strategy described above leads to the determination of both material parameters and domains of validity corresponding to the different loading conditions for each model. Moreover, in regards to some criteria, a classification of the models is proposed in the following.

So, the previous fitting procedure is applied to all models described. The corresponding material parameters are given in Tables II and III respectively for phenomenological (here, the term 'phenomenological' stands for models founded on mathematical developments and also experimental determination of $(\partial W/\partial I_i)_{i=1,2}$ which were presented separately above) and physically-based constitutive models. Lines indexed by (T) correspond to parameters obtained with Treloar experiments and lines indexed by (K) to those obtained with Kawabata *et al.* ones. In these tables, the unit is MPa for pressure parameters. In fact, if the (T) and (K) parameters are equal, it means that the same set of parameters is able to fit the data of Treloar and Kawabata *et al.* simultaneously.

In order to illustrate the present results, some comparisons between experimental and predicted stress-strain data are given in Appendix.

RANKING

Finally, the previous work is used to propose a ranking of the twenty hyperelastic models investigated here. This ranking is established in regards with the ability of the models to reproduce two given sets of experimental data, i.e. those of Treloar[14] and Kawabata *et al.*[59], obtained with two similar unfilled natural rubbers. Nevertheless, considering the various loading conditions covered by these two sets, results can be extended to other elastomers and the following ranking should be seen as a decision tool for engineers who deal with finite element simulation of rubber parts.

The ranking is based on the following rules :

- First, larger is the validity range of a model for the complete behavior (different types of loading conditions), upper is ranked this model.
- Then, greater is the number of material parameters of a model, lower is ranked this model.
- Moreover, for equivalent models in regards with the two previous rules, the one which is able to reproduce both experimental data sets with the same set of material parameters is considered as the best.
- Finally, a more subjective rule is adopted to separate equivalent models in regards to the three first rules. Award is delivered to physically-based models. In fact, this final rule is justified when a hyperelastic formulation is used as the basis of the development of inelastic constitutive equations (viscoelasticity, Mullins effect ...). Indeed, if the material parameters are physically motivated, their time evolution can also be predicted by physical observation and this can be used to defined evolution laws for inelastic models (see for example the case of the Mullins effect in[5]).

So with these rules, the ranking of the 20 hyperelastic models is established and given in Table IV.

TABLE II: — PARAMETERS OF PHENOMENOLOGICAL HYPERELASTIC MODELS: (T) FOR TRELOAR DATA, (K) FOR KAWABATA ET AL. DATA

Data	Model name Fitted parameters
	Mooney
(T)	$C_1 = 0.162$, $C_2 = 5.90\ 10^{-3}$
(K)	$C_1 = 0.182$, $C_2 = 9.79\ 10^{-3}$
	Biderman
(T)	$C_{10} = 0.208$, $C_{01} = 2.33\ 10^{-2}$, $C_{20} = -2.40\ 10^{-3}$, $C_{30} = 5\ 10^{-4}$
(K)	$C_{10} = 0.185$, $C_{01} = 1.27\ 10^{-2}$, $C_{20} = -2.90\ 10^{-3}$, $C_{30} = 1.77\ 10^{-5}$
	Haines-Wilson
(T)	$C_{10} = 0.173$, $C_{01} = 6.68\ 10^{-3}$, $C_{11} = -1.18\ 10^{-4}$, $C_{20} = -1.19\ 10^{-3}$, $C_{02} = 2.3\ 10^{-6}$, $C_{30} = 3.85\ 10^{-5}$
(K)	$C_{10} = 0.176$, $C_{01} = 2.34\ 10^{-2}$, $C_{11} = -1.17\ 10^{-3}$, $C_{20} = -4.64\ 10^{-3}$, $C_{02} = 1.59\ 10^{-5}$, $C_{30} = 2.47\ 10^{-4}$
	Gent and Thomas
(T)	$C_1 = 0.176$, $C_2 = 5.65\ 10^{-2}$
(K)	$C_1 = 0.153$, $C_2 = 0.147$
	Hart-Smith
(T)	$G = 0.175$, $k_1 = 2.86\ 10^{-4}$, $k_2 = 0.311$
(K)	$G = 0.145$, $k_1 = 8.42\ 10^{-4}$, $k_2 = 1.13\ 10^{-4}$
	Valanis and Landel
(T)	$\mu = 0.449$
(K)	$\mu = 0.418$
	Ogden
(T)	$\alpha_1 = 1.3$, $\mu_1 = 0.63$, $\alpha_2 = 5$, $\mu_2 = 1.2\ 10^{-3}$, $\alpha_3 = -2$, $\mu_3 = -1\ 10^{-2}$
(K)	$\alpha_1 = 1.37$, $\mu_1 = 0.54$, $\alpha_2 = 3.91$, $\mu_2 = 5.19\ 10^{-3}$, $\alpha_3 = -1.56$, $\mu_3 = -2.15\ 10^{-2}$
	Gent
(T)	$E = 0.978$, $I_m = 96.4$
(K)	$E = 1.19$, $I_m = 22.7$
	Yeoh and Fleming
(T)	$A = 0.0519$, $B = 4.03$, $C_{10} = 1.127$, $I_m = 82.8$
(K)	$A = 0.0251$, $B = 31.1$, $C_{10} = 0.179$, $I_m = 42.3$
	Shariff
(T)	$E = 1.072$, $\alpha_1 = 0.896$, $\alpha_2 = 3.98\ 10^{-2}$, $\alpha_3 = 8.88\ 10^{-5}$, $\alpha_4 = 2.73\ 10^{-2}$
(K)	$E = 1.072$, $\alpha_1 = 0.896$, $\alpha_2 = 3.98\ 10^{-2}$, $\alpha_3 = 8.88\ 10^{-5}$; $\alpha_4 = 2.73\ 10^{-2}$

TABLE III: — PARAMETERS OF PHYSICALLY-BASED HYPERELASTIC MODELS: (T) FOR TRELOAR DATA, (K) FOR KAWABATA ET AL. DATA

Data	Model name Fitted parameters
	neo-Hookean
(T)	$\frac{1}{2}nkT = 0.2$
(K)	$\frac{1}{2}nkT = 0.2$
	3-chains
(T)	$\frac{1}{2}nkT = 0.283$, $N = 75.9$
(K)	$\frac{1}{2}nkT = 0.356$, $N = 365$
	Ishihara
(T)	$C_{10} = 0.171$, $C_{01} = 4.89\ 10^{-3}$, $C_{20} = -2.4\ 10^{-4}$
(K)	$C_{10} = 0.186$, $C_{01} = 1.04\ 10^{-2}$, $C_{20} = 2.52\ 10^{-3}$
	slip-link
(T)	$N_ckT = 0.3$, $N_skT = 0.53$, $\eta = 1.9$
(K)	$N_ckT = 0.31$, $N_skT = 0.29$, $\eta = 0.84$
	van der Waals
(T)	$G = 0.434$, $a = 0.320$, $\lambda_m = 10.24$, $\beta = 0.958$
(K)	$G = 0.417$, $a = 0.303$, $\lambda_m = 10.1$, $\beta = 0.93$
	constrained junctions
(T)	$C_{10} = 0.16$, $\frac{1}{2}kT\mu = 0.7$, $\kappa = 1.55$
(K)	$C_{10} = 0.166$, $\frac{1}{2}kT\mu = 0.5$, $\kappa = 1.7$
	8-chains
(T)	$C_r = 0.28$, $N = 25.4$
(K)	$C_r = 0.394$, $N = 45.4$
	tube
(T)	$G_c = 0.266$, $G_e = 0.111$, $\beta = 0.375$
(K)	$G_c = 0.266$, $G_e = 0.111$, $\beta = 0.375$
	extended-tube
(T)	$G_c = 0.202$, $G_e = 0.153$, $\beta = 0.178$, $\delta = 0.0856$
(K)	$G_c = 0.202$, $G_e = 0.153$, $\beta = 0.178$, $\delta = 0.0856$
	micro-sphere
(T)	$\mu = 0.292$, $N = 22.01$, $p = 1.472$, $U = 0.744$, $q = 0.1086$
(K)	$\mu = 0.292$, $N = 22.01$, $p = 1.472$, $U = 0.744$, $q = 0.1086$

TABLE IV: — RANKING OF 20 HYPERELASTIC MODELS FOR RUBBERLIKE MATERIALS. SEE THE SIGNIFICANCE OF COLUMNS IN THE TEXT

						Treloar data λ_{max}				Kawabata *et al.* data λ_{max}	
	Model	Year	N.m.p.	Phys		UE	PS	EQE	BE	λ_1	λ_2
1	extended-tube	1999	4	×	=	-	-	-	-	-	-
2	Shariff	2000	5		=	-	-	-	-	-	-
3	micro-sphere	2004	5	×	=	-	-	-	-	-	-
4	Ogden	1972	6		≠	-	-	-	-	-	-
5	Haines-Wilson	1975	6		≠	-	-	-	-	3.4	3
6	Biderman	1958	4		≠	-	-	-	-	2.5	3
7	Hart-Smith	1966	3		=	-	-	-	-	1.9	1.5
8	8-chain	1993	2	×	≠	-	-	under	under	1.9	1.9
9	Gent	1996	2		≠	-	-	-	-	1.6	1.6
10	Yeoh and Fleming	1997	4		≠	-	-	-	-	1.6	1.6
11	van der Waals	1986	4	×	=	-	-	2.5	over	2.2	2.2
12	3-chain	1943	2	×	≠	-	-	under	under	1.3	1.3
13	tube	1997	3	×	=	4	3.5	3	-	-	-
14	Mooney	1940	2		≠	5	-	4	2	2.2	2
15	Ishihara	1951	3	×	≠	5	-	4	2.25	1.9	1.9
16	Gent and Thomas	1958	2		=	5	-	3	-	1.6	1.6
17	slip-link	1981	3	×	≠	5	4	2.5	over	2.5	2.5
18	constrained junctions	1982	3	×	≠	5	4	2.5	over	2.2	2.2
19	neo-Hookean	1943	1	×	=	5	2	3	2.5	1.6	1.6
20	Valanis and Landel	1967	1		≠	3.5	2.5	1.2	under	1.3	1.3

In Table IV, models are classified from the best to the worst. The first column contains the rank, the second the name of the model. Then the three following columns contain informations given above but recalled here: the year of development, the number of material parameters (N.m.p.) and if the model is physically-based ($\times$) or not (nothing). Then, the symbol in the sixth column means that the same set of material parameters is able to fit simultaneously the experimental data of Treloar and Kawabata *et al.* ($=$) or not ($\neq$). Columns 7-10 summarize the results obtained with Treloar data. For each type of loading conditions (UE, PS, EQE, BE), the validity range of the model is given in terms of stretch: a dash (-) means that the model is efficient for the whole range of experimental stretches, a number represents the upper limit of the validity range, and terms "under" and "over" signify that the stress is respectively underestimated or overestimated. The two last columns give similar information for the data set of Kawabata *et al.*: they define the validity range in terms of the maximum stretches in the two loading directions, λ_1 and λ_2.

FINAL REMARKS

This ranking leads to some remarks. First, only four models are revealed able to fit all experimental data considered here: the extended-tube[54], Shariff[31], micro-sphere[55] and Ogden[19] models. Among them, only the first three ones admit the same material parameters for both data sets. These three models are recent and they are not widely used in industrial context. The best model is the extended-tube model because it involves only four parameters and its derivation is physically-motivated. The Ogden model is older and is classically used for finite element simulations. It is quite efficient but its six material parameters necessitate a large experimental database to be fitted.

Second, it is highlighted that models with only two or three material parameters are unable to predict the whole range of strain, even if they are derived for large strain response. This is the case of the 3-chain[39], Hart-Smith[66], 8-chain[35] and Gent[34] models. Their inefficiency is revealed for predicting the biaxial response of rubber if their parameters are determined with uniaxial data.

Third, for moderate strain, i.e. 200-250%, the "old" Mooney model[22] (two material parameters) is the most efficient. Indeed, physically-motivated models, such as the slip-link[45], van der Waals[8,46–48], constrained junctions[40,43,49,50] and tube[51] models, involve three parameters and their abilities to predict moderate strain response are quite similar to the one of the Mooney model.

For small strain, i.e. about 150% and below, the neo-Hookean constitutive equation[36] should be used for three reasons: it is physically-founded even if the basic assumptions are quite simplistic, it involves only one material parameter and it is able to predict the material response for different types of loading conditions (the same value of the parameter was obtained for both experimental data sets).

APPENDIX

In this appendix, the efficiency of eight models is illustrated by comparing their response to experimental data which were used for fitting.

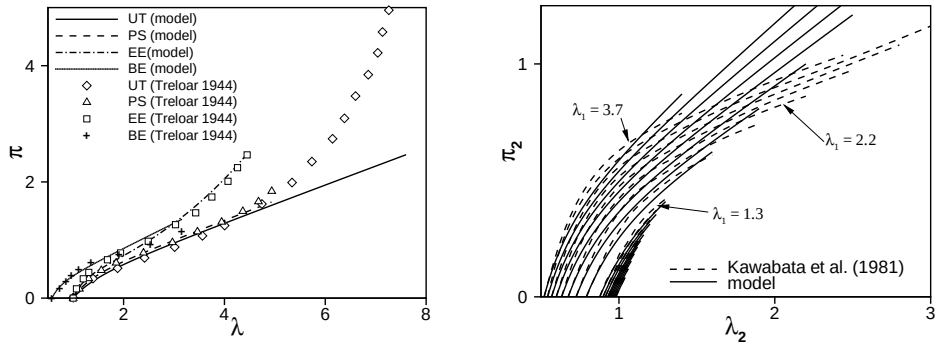

FIG. 1. : — Comparison between the prediction of the Mooney model and the experimental data of Treloar (left-hand side graph) and Kawabata *et al.* (right-hand side graph)

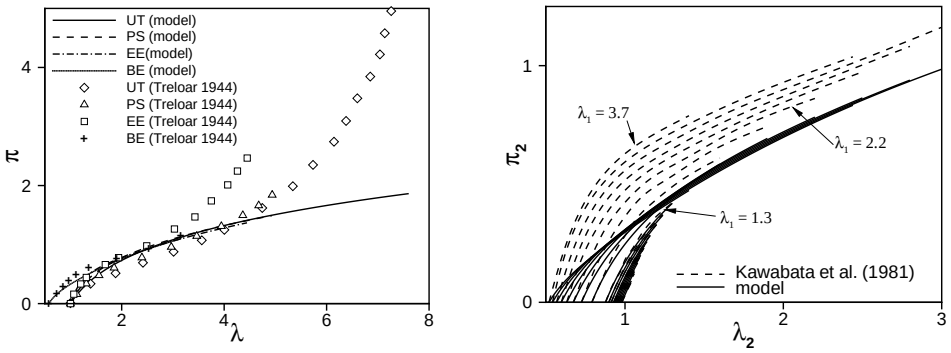

FIG. 2. : — Comparison between the prediction of the Valanis and Landel model and the experimental data of Treloar (left-hand side graph) and Kawabata *et al.* (right-hand side graph)

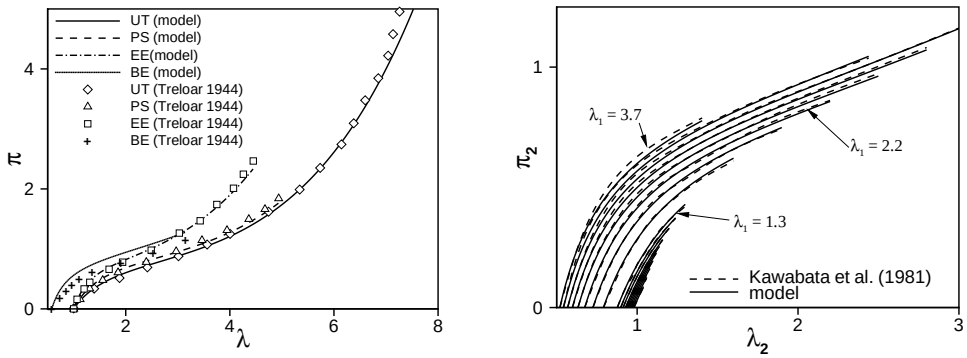

FIG. 3. : — Comparison between the prediction of the Ogden model and the experimental data of Treloar (left-hand side graph) and Kawabata *et al.* (right-hand side graph)

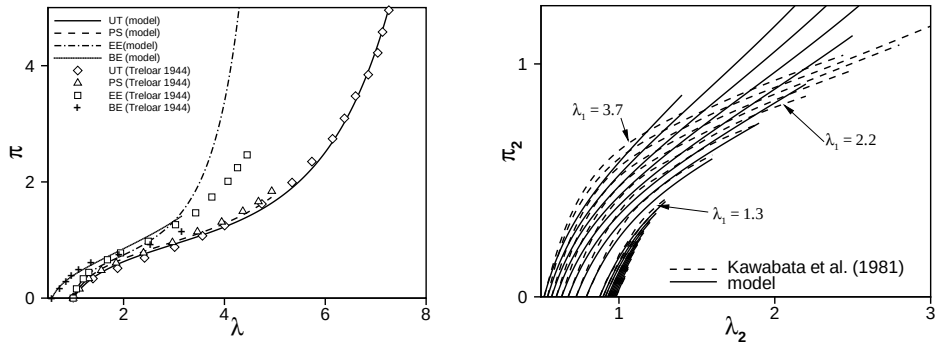

FIG. 4. : — Comparison between the prediction of the van der Waals model and the experimental data of Treloar (left-hand side graph) and Kawabata *et al.* (right-hand side graph)

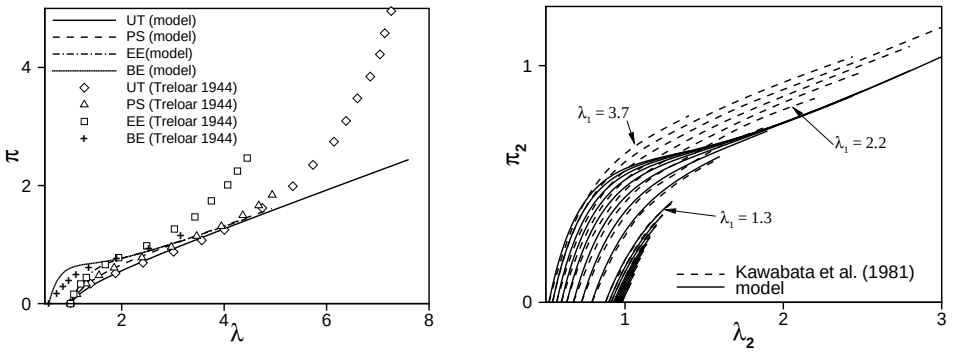

FIG. 5. : — Comparison between the prediction of the constrained junctions model and the experimental data of Treloar (left-hand side graph) and Kawabata *et al.* (right-hand side graph)

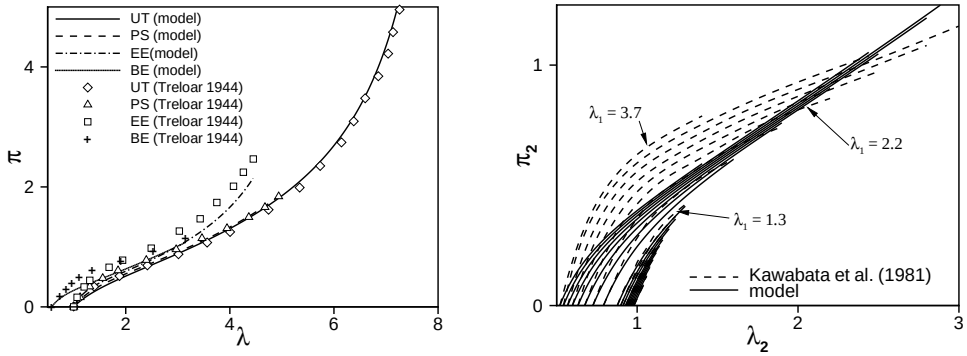

FIG. 6. : — Comparison between the prediction of the 8-chains model and the experimental data of Treloar (left-hand side graph) and Kawabata *et al.* (right-hand side graph)

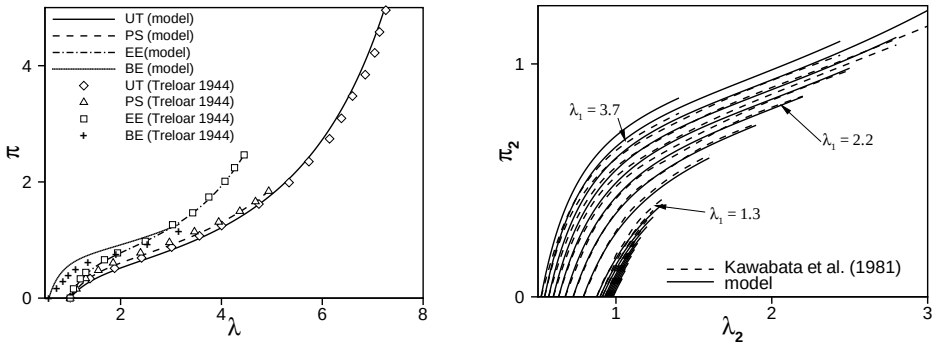

FIG. 7. : — Comparison between the prediction of the extended-tube model and the experimental data of Treloar (left-hand side graph) and Kawabata *et al.* (right-hand side graph)

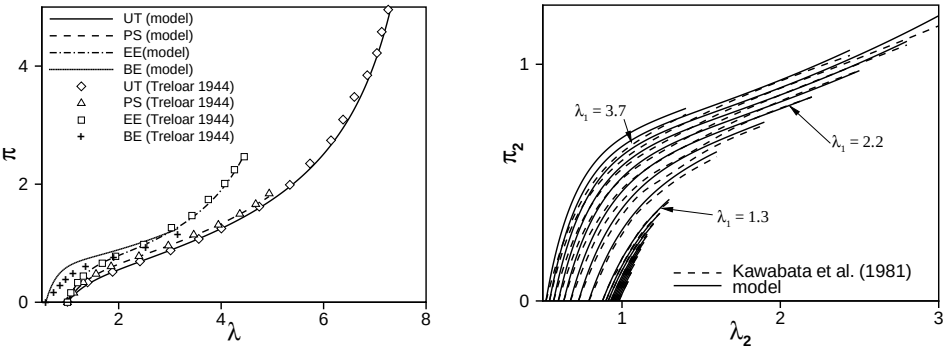

FIG. 8. : — Comparison between the prediction of the micro-sphere model and the experimental data of Treloar (left-hand side graph) and Kawabata *et al.* (right-hand side graph)

REFERENCES

[1] Gent, A. N. "Engineering With Rubber: How to Design Rubber Components", Hanser Gardner, 2nd ed. edition, (2001).

[2] Ogden, R. W. and Roxburgh, D. G. *Proc. R. Soc. Lond. A* **455**, 2861–2877 (1999).

[3] Miehe, C. and Keck, J. *J. Mech. Phys. Solids* **48**, 323–365 (2000).

[4] Bergström, J. S. and Boyce, M. C. *Mech. Mat.* **33**, 523–530 (2001).

[5] Marckmann, G., Verron, E., Gornet, L., Chagnon, G., Charrier, P., and Fort, P. *J. Mech. Phys. Solids* **50**, 2011–2028 (2002).

[6] Drozdov, A. D. and Dorfmann, A. *Meccanica* **39**, 245–270 (2004).

[7] Dorfmann, A. and Ogden, R. *Int. J. of Solids Struct.* **41**(7), 1855–1878 April (2004).

[8] Kilian, H. G., Enderle, H. F., and Unseld, K. *Colloid Polym. Sci.* **264**, 866–879 (1986).

[9] Yeoh, O. H. and Fleming, P. D. *J. Polymer Sci. Part B: Polymer Phy.* **35**, 1919–1931 (1997).

[10] Boyce, M. C. Rubber Chem. Technol. **69**, 781–785 (1996).

[11] Chagnon, G., Marckmann, G. and Verron, E. Rubber Chem. Technol. **77**, 724–735 (2004).

[12] Seibert, D. J. and Schöche, N. Rubber Chem. Technol. **73**, 366–384 (2000).

[13] Boyce, M. C. and Arruda, E. M. Rubber Chem. Technol. **73**, 505–523 (2000).

[14] Treloar, L. R. G. *Trans. Faraday Soc.* **40**, 59–70 (1944).

[15] Attard, M. M. and Hunt, G. W. *Int. J. Solids Struct.* **41**, 5327–5350 (2004).

[16] Bonet, J. and Wood, R. D. "Nonlinear Continuum Mechanics for Finite Element Analysis", Cambridge University Press, Cambridge, (1997).

[17] Holzapfel, G. A. "Nonlinear Solid Mechanics. A continuum approach for engineering", J. Wiley and Sons, Chichester, (2000).

[18] Rivlin, R. S. *Philos. T. Roy. Soc. A* **240**, 459–490 (1948).

[19] Ogden, R. W. *Proc. R. Soc. Lond. A* **326**, 565–584 (1972).

[20] Rivlin, R. S. and Saunders, D. W. *Philos. T. Roy. Soc. A* **243**, 251–288 (1951).

[21] Hart-Smith, L. J. *Z. angew. Math. Phys.* **17**, 608–626 (1966).

[22] Mooney, M. *J. Appl. Phys.* **11**, 582–592 (1940).

[23] Rivlin, R. S. *Philos. T. Roy. Soc. A* **241**, 379–397 (1948).

[24] Tschoegl, N. W. *J. Polymer Sci. Part A-1* **9**, 1959–1970 (1971).

[25] James, A. G., Green, A., and Simpson, G. M. *J. Appl. Polym. Sci.* **19**, 2033–2058 (1975).

[26] Yeoh, O. H. Rubber Chem. Technol. **63**(5), 792–805 (1990).

[27] Criscione, J. C., Humphrey, J. D., A.S., A. S. D., and Hunter, W. C. *J. Mech. Phys. Solids* **48**, 2445–2465 (2000).

[28] Biderman, V. L. *Rascheti na Prochnost* **40**, (1958).

[29] Alexander, H. *Int. J. Engrg. Sci.* **6**, 549–562 (1968).

[30] Hill, R. *Adv. Appl. Mech.* **18**, 1–75 (1978).

[31] Shariff, M. H. B. M. Rubber Chem. Technol. **73**, 1–18 (2000).

[32] Gent, A. N. and Thomas, A. G. *J. Polym. Sci.* **28**, 625–637 (1958).

[33] Valanis, K. C. and Landel, R. F. *J. Appl. Phys.* **38**(7), 2997–3002 (1967).

[34] Gent, A. N. Rubber Chem. Technol. **69**, 59–61 (1996).

[35] Arruda, E. and Boyce, M. C. *J. Mech. Phys. Solids* **41**(2), 389–412 (1993).

[36] Treloar, L. R. G. *Trans. Faraday Soc.* **39**, 36–64 ; 241–246 (1943).

[37] Treloar, L. R. G. "The physics of rubber elasticity", Oxford University Press, Oxford (UK), (1975).

[38] Kuhn, W. and Grün, F. *Kolloideitschrift* **101**, 248–271 (1942).

[39] James, H. M. and Guth, E. *J. Chem. Phys.* **11**, 455–481 (1943).

[40] Flory, P. J. *Chem. Rev.* **35**, 51–75 (1944).

[41] Treloar, L. R. G. *Trans. Faraday Soc.* **42**, 77–83 (1946).

[42] Isihara, A., Hashitsume, N., and Tatibana, M. *J. Chem. Phys.* **19**, 1508–1512 (1951).

[43] Erman, B. and Flory, P. *Macromolecules* **15**, 806–811 (1982).

[44] Mark, J. E. and Erman, B. "Rubberlike elasticity - A molecular primer", J. Wiley and Sons, New-York, (1988).

[45] Ball, R. C., Doi, M., Edwards, S. F., and Warner, M. *Polymer* **22**, 1010–1018 August (1981).

[46] Kilian, H. G. *Polymer* **22**, 209–217 (1981).

[47] Enderle, H. F. and Kilian, H. G. *Prog. Colloid Polym. Sci.* **75**, 55–61 (1987).

[48] Ambacher, H., Enderle, H. F., Kilian, H. G., and Sauter, A. *Prog. Colloid Polym. Sci.* **80**, 209–220 (1989).

[49] Flory, P. J. and Erman, B. *Macromolecules* **15**, 800–806 (1982).

[50] Flory, P. J. *Chem. Rev.* **35**, 51–75 (1994).

[51] Heinrich, G. and Kaliske, M. *Comput. Theo. Polym. Sci.* **7**(3/4), 227–241 (1997).

[52] Edwards, S. F. and Vilgis, T. A. *Rep. Prog. Phys.* **51**, 243–297 (1988).

[53] Doi, M. "Introduction to Polymer Physics", Oxford Science Publications, Oxford, (1996).

[54] Kaliske, M. and Heinrich, G. Rubber Chem. Technol. **72**, 602–632 (1999).

[55] Miehe, C., Goktepe, S., and Lulei, F. *J. Mech. Phys. Solids* **52**, 2617–2660 (2004).

[56] Treloar, L. R. G. and Riding, G. *Proc. R. Soc. Lond. A* **369**, 261–280 (1979).

[57] Wu, P. D. and van der Giessen, E. *J. Mech. Phys. Solids* **41**(3), 427–456 (1993).

[58] Bazant, Z. P. and Oh, B. H. *Z. angew. Math. Mech.* **66**(1), 37–49 (1986).

[59] Kawabata, S., Matsuda, M., Tei, K., and Kawai, H. *Macromolecules* **14**, 154–162 (1981).

[60] Lambert-Diani, J. and Rey, C. *J. Phys. IV* **9**, 117–126 (1999).

[61] Criscione, J. C. *J. Elasticity* **70**, 129–147 (2003).

[62] Charlton, D. J., Yang, J., and Teh, K. K. Rubber Chem. Technol. **67**, 481–503 (1994).

[63] Marckmann, G. "Contribution à l'étude des élastomères et des membranes soufflées", PhD Thesis, Ecole Doctorale Mécanique, Thermique et Génie Civil, Ecole Centrale de Nantes et Université de Nantes, 07 juin (2004).

[64] Carmichael, A. J. and Holdaway, H. W. *J. Appl. Phys.* **32**(2), 159–166 (1961).

[65] Klingbeil, W. W. and Shield, R. T. *Z. angew. Math. Phys.* **15**, 608–629 (1964).

[66] Hart-Smith, L. J. and Crisp, J. D. C. *Int. J. Engrg. Sci.* **5**, 1–24 (1967).

[67] Holland, J. H. "Adaptation in natural and artificial systems", University of Michigan Press, Ann Arbor, (1975).

[68] Goldberg, D. E. "Algorithmes génétiques, exploitation, optimisation et apprentissage automatique", Addison-Wesley eds., (1994).

[69] Furukawa, T. and Yagama, G. *Int. J. Num. Meth. Engrg.* **40**, 1071–1090 (1997).

[70] Liu, G., Han, X., and Lam, K. *Comp. Meth. Appl. Mech. Engrg.* **191**, 1909–1921 (2002).

[71] Yoshimoto, F., Harada, T., and Yoshimoto, Y. *Comp. Aided Design* **35**, 751–760 (2003).

[72] Levenberg, K. *Quart. Appl. Math.* **2**, 164–168 (1944).

[73] Marquardt, D. W. *J. Soc. Indust. Appl. Math.* **11**(2), 431–441 (1963).

[74] Culioli, J.-C. "Introduction à l'optimisation", Ellipses, Ecole des Mines de Paris, (1994).