

1 **Mokarram Hossain · A. F. M. S. Amin · Muhammad Nomani**

2 **Kabir**

3 **Eight-chain and full-network models and their modified**  
4 **versions for rubber hyperelasticity : A comparative study**

5  
6 Received: date / Accepted: date

7 **Abstract** The eight-chain model (Arruda EM, Boyce MC, 1993. A three-dimensional constitutive model  
8 for the large stretch behaviour of rubber elastic materials. J. Mech. Phy. Solids 41, 389-412), also known  
9 as Arruda-Boyce model, is widely used to capture the rate-independent hyperelastic response of rubberlike  
10 materials. The parameters of this model are physically based and explained from micromechanics of chain  
11 molecules. Despite its excellent performance with only two material parameters to capture bench measure-  
12 ments in uniaxial and pure shear regime, the model is known to be significantly deficient in predicting the  
13 equibiaxial data. To ameliorate such drawback, over the years, several modified versions of this successful  
14 model have been proposed in the literature. The so-called full-network model is another micromechanically-  
15 motivated chain model, which has also few modified versions in the literature. For this study, two modified  
16 versions of the full-network model have been selected. In this contribution, five modified versions of the  
17 Arruda-Boyce model and two modified versions of full-network model are critically compared with the clas-  
18 sical eight chain model for their adequacy in representing equibiaxial data. To do a comparison of all selected  
19 models in reproducing the well-known Treloar data (Trans Faraday Soc 40:59-70, 1944), the analytical ex-  
20 pressions for the three homogeneous deformation modes, i.e. uniaxial tension, equibiaxial tension and pure  
21 shear have been derived and the performances of the selected models are analysed. The comparative study

---

Mokarram Hossain  
Chair of Applied Mechanics, University of Erlangen-Nuremberg, Germany  
Tel.: +49-9131-8528514  
Fax: +49-9131-8528503  
E-mail: mokarram.hossain@lrm.uni-erlangen.de

A. F. M. S. Amin  
Department of Civil Engineering, Bangladesh University of Engineering and Technology, Dhaka, Bangladesh  
Tel.: +880-2-8616833, Ext 7944  
E-mail: samin@ce.buet.ac.edu

Muhammad Nomani Kabir  
Department of Computer Science, Taibah University, Madinah, Saudi Arabia  
E-mail: dr.nomankabir@gmail.com

---

1 demonstrates that modified Flory-Erman model, Gornet-Desmorat (GD) model, Meissner-Matějka model and  
2 Bootstrapped eight-chain model predict well the three deformation modes compare to the classical eight-chain  
3 model.

4 **Keywords** Eight-chain model · Full-network model · Rubber-like material · Phenomenological model ·  
5 Micromechanical model

## 6 **1 Introduction**

7 Rubber-like materials have enormous applications especially in industrial and engineering fields, such as  
8 tires, engine mounts, seals, conveyor belts, base isolations for protecting buildings and bridges from devas-  
9 tating earthquakes [1; 2; 3; 4; 37]. Such wide-range of applications of rubbers make the rubber-mechanics as  
10 one of the major active fields of research in last several decades. Especially the fast growing numerical tech-  
11 niques such as the finite element method facilitate sophisticated design and analysis for complex large strain  
12 three-dimensional elastomeric components. In the development of numerical models, a simple but advanced  
13 constitutive model to capture multi-axial data is an essential ingredient. The large deformability together  
14 with recoverability of rubber-like materials are well known for their highly nonlinear load-deformation be-  
15 haviour. These materials are generally modelled by considering homogeneous, isotropic, incompressible or  
16 nearly incompressible, geometrically and physically nonlinear (visco) elastic solids. Such idealizations are  
17 also supported by experimental data [15; 27].

18 In the literature [9; 26], elastomeric material models are generally classified into two main categories: purely  
19 *phenomenological* and *micromechanical* based network models. The micromechanically-motivated network  
20 models, on the one hand, are based on the statistics of cross-linked long chain molecules [9] and the phe-  
21 nomenological models, on the other hand, involve invariant or principal stretch-based macroscopic contin-  
22 uum formulations generally having polynomial structures. The governing parameters appearing in the pro-  
23 posed expressions of the energy functions of the phenomenological models do not have, in most of the  
24 cases, any physical interpretation. The three-chain, four-chain, eight-chain, full network models, tube-model,  
25 extended-tube model, Flory-Erman model, micro-macro unit sphere model, Kroon model are well-known  
26 micromechanically-inspired models, which can be used for moderate to large elastic deformations of poly-  
27 meric materials, see for example, Boyce and Arruda [9], Arruda and Boyce [11], Miehe et al. [8], Hossain  
28 and Steinmann [26]. Some well-known phenomenological models are Neo-Hooke, Mooney-Rivlin, Ogden,  
29 Gent, Yeoh, Attard, Shariff, Hart-Smith, van der Waals, Lopez-Pamies, Pucci-Saccomandi, Carroll etc, for  
30 more details, see Hossain and Steinmann [26], Steinmann et al. [28].

31 As mentioned earlier, numerous constitutive models have been proposed during the last several decades to  
32 describe the elastic response of elastomers, but only a few of them are able to satisfactorily reproduce ex-

---

1 experimental data for different deformation modes, i.e. uniaxial and biaxial extensions, simple and pure shears.  
2 According to Marckmann and Verron [16], the promising candidate for the best model will be that one,  
3 which can describe the complete behaviour of elastomers with a minimal number of material parameters that  
4 can be determined from explicit experimental data without facing any difficulty, e.g. instability. Marckmann  
5 and Verron [16], after comparing twenty models, placed the extended tube model [17], Shariff model [23],  
6 micro-macro unit sphere model [8] and Ogden [12; 13] model at the top of their ranking list. Earlier, Boyce  
7 and Arruda [9] conducted an excellent review on several hyperelastic models where they utilized Treloar's  
8 experimental data for comparison of three type of deformations (uniaxial, biaxial and pure shear).

9 The eight-chain model is a classical micromechanically-motivated constitutive model for polymeric materials,  
10 where the governing parameters have direct links to the micro-mechanics of the polymer molecular structures.  
11 With only two material constants, this model excellently captures major classical experimental data available  
12 in the literature. Despite its excellent performance to capture the uniaxial and pure shear data in the case of  
13 Treloar, the model prediction faces a significant deficiency to calibrate the equibiaxial data [9; 32]. Several  
14 modified versions of this model were proposed over the last two decades to show a better performance in the  
15 equibiaxial range. Some variants are of structural types while the extended or modified parts of some other  
16 variants are based purely on phenomenological consideration, cf. [30]. For the sake of modifications over the  
17 classical one, some models increase the number of parameters to fit the data, e.g. Bechir model [33] while  
18 some remain with the two parameters, e.g. Bootstrapped eight-chain model [32]. Some well-known modified  
19 versions of the Arruda-Boyce model are: modified Flory-Erman model [9; 21], Gornet-Desmorat (GD) model  
20 [30], Bootstrapped eight-chain model [32], Bechir model [33], Meissner-Matejka model [34], Kroon model  
21 [15]. When a modified model is proposed aiming to show better results in contrast to the original eight-chain  
22 model, the modified version is usually compared with the original one but the comparison among the variants  
23 of the eight-chain model are not demonstrated so far in the literature. Therefore, such comparative study can  
24 work as a guideline for a beginner who wants to work in rubber mechanics to choose an appropriate model  
25 from several variants.

26 The full-network model is also a micromechanically-inspired model where polymer chains are assumed to be  
27 randomly oriented in space. Due to the computational cost for the integration over a sphere, either analytically  
28 or numerically, several simplified approaches for this model have been proposed in the literature [14; 24; 36]  
29 where analytical or numerical integration over the sphere is bypassed. For this study, we have selected the  
30 modified full-network model due to Wu and Giessen [14] and the another one that is proposed in Zuniga and  
31 Beatty [24]. The aim of the current study is to present a comparative study among all modified versions so far  
32 available in the literature, at least to the best of the author's knowledge.

33 The paper is structured as follows: in Section 2 we briefly review the general framework for the derivation  
34 of (semi) analytical stress-strain relations from arbitrary free energy functions for the homogeneous cases. In

1 Section 3, the analytical formulations for the three homogeneous deformation modes, i.e. uniaxial tension,  
 2 equibiaxial tension and pure shear are derived for all selected models. These are necessary to evaluate the  
 3 performance of all chosen models in reproducing the experimental data provided by Treloar. To this end a  
 4 standard fitting tool is applied to calculate the optimal model parameters with respect to each set of Treloar's  
 5 data. Section 3 also presents a comparative study on six modified models along with the original eight-chain  
 6 and full-network models. Finally concluding remarks close the paper.

## 7 **2 Analytical formulations**

Concerning comparison of a novel model with the existing one (s), most of the authors used constitutive  
 (semi) analytical expressions and the classical Treloar data. Since these data are given in pairs of principal  
 stretches  $\lambda_i$  and principal 1<sup>st</sup>-Piola-Kirchhoff (PK) stresses  $P_i$  for different deformation modes, e.g. uniaxial  
 tension (UN), equibiaxial tension (EB) and pure shear (PS), the analytical formulations for the principal 1<sup>st</sup>-  
 Piola-Kirchhoff (PK), i.e.  $P_i(\lambda_i)$  need to be derived from the particular free energy function. Then, in order  
 to compare the analytical expression with the experimental data, an optimal set of material parameters for  
 each model and a particular deformation mode has to be identified. For the parameter identification, different  
 optimization techniques can be used. The material investigated by Treloar is characterised as isotropic and  
 incompressible, a case for which principal stretches and 1<sup>st</sup>-PK stresses are related by

$$P_i = \frac{\partial \Psi}{\partial \lambda_i} - \frac{1}{\lambda_i} p \quad , \quad i = 1, 2, 3 \text{ (no sum)} \quad (1)$$

where  $p$  denotes the hydrostatic pressure that has to be determined from appropriate boundary conditions, for  
 details see [9; 26]. For the case of invariant based models, i.e.  $\Psi = \tilde{\Psi}(I_1, I_2)$ , the equation (1) leads to

$$P_i = \frac{\partial \Psi}{\partial I_1} \frac{\partial I_1}{\partial \lambda_i} + \frac{\partial \Psi}{\partial I_2} \frac{\partial I_2}{\partial \lambda_i} - \frac{1}{\lambda_i} p \quad , \quad i = 1, 2, 3 \text{ (no sum)}. \quad (2)$$

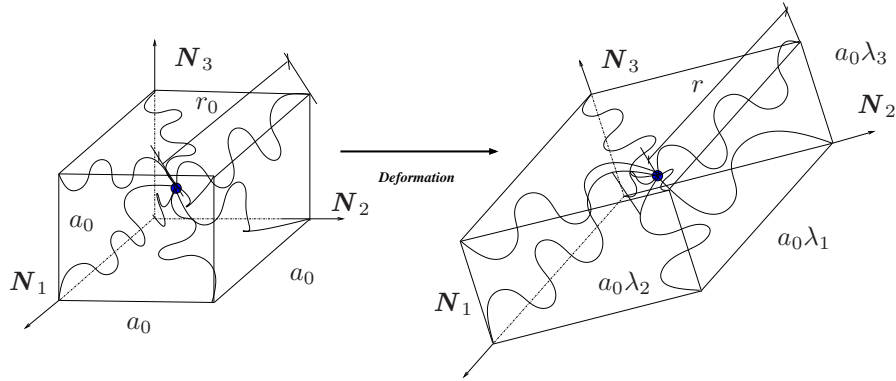
8 Using Eqns (1) and (2) we derive the required analytical formulations for the three homogeneous deformation  
 9 modes. For further details, see Hossain and Steinmann [26], Steinmann et al. [28]. To identify the material  
 10 parameter set for each model, an open source computer code, TRESNEI is used. The code is suitable for  
 11 bound-constrained non-linear least-square problems, cf. [29]. For the validation of each model, each set of  
 12 optimal material parameters for UN, EB and PS is used to compute the response of the other two deformation  
 13 modes. Each subsection contains the corresponding figures that also contain the errors between (a) each  
 14 experiment and its optimal fit, e.g. Error(UN-fit), and (b) the simulations of the other deformation modes and  
 15 their respective measurements, e.g. Error(EB-sim). All error calculations have been performed according to  
 16 the relation below

$$\text{Error}(\text{fit}/\text{sim}) = \sqrt{\frac{1}{M} \sum_{i=1}^M \left[ P_{\text{fit}/\text{sim}}(\lambda_i^{\text{Treloar}}) - P_{\text{Treloar}}(\lambda_i^{\text{Treloar}}) \right]^2}, \quad (3)$$

- 1 i.e. sum up the squared differences between fitted/simulated and measured first Piola-Kirchhoff stresses, re-  
 2 spectively, averaged by the number of data points  $M$  (stretches) available for each deformation mode.

### 3 Comparative study: Model performances

#### 4 3.1 Eight-chain model



**Fig. 1** Eight-chain model: initial and deformed chain orientations and stretches

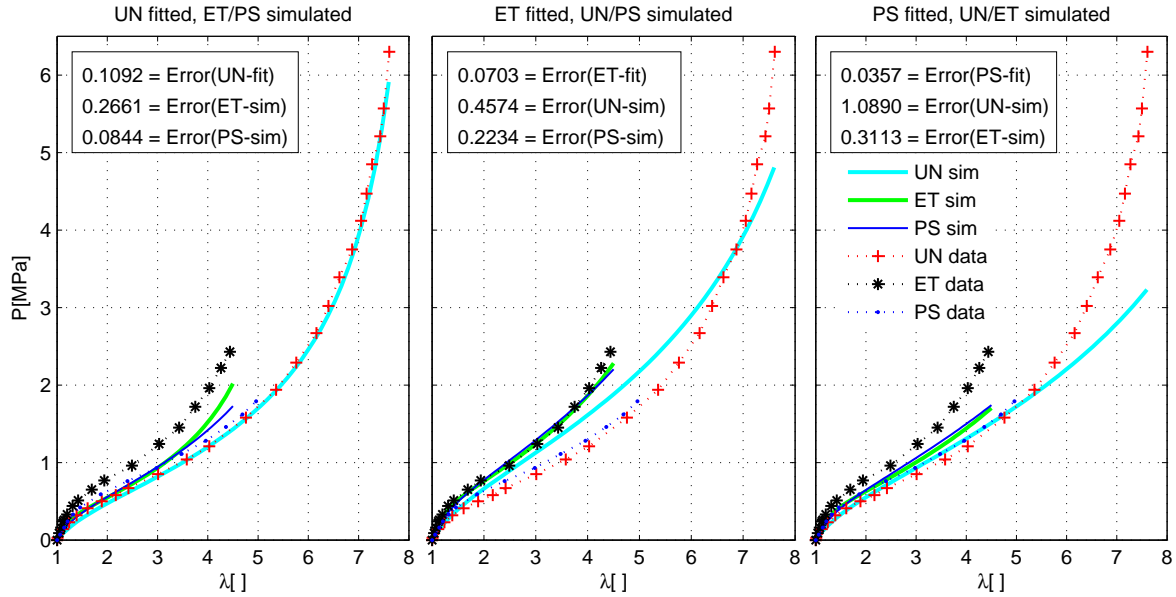
In this model, the cuboid volume element, cf. Fig (1) is assumed to have edges parallel to the principal directions, which is composed of eight chains (hence the name eight-chain model) oriented in the diagonals from the center of the volume to its corners. The relation between edge length  $a_0$  of the cuboid and the chain length  $r_0$ , in the undeformed state, is given by simple geometric relations,  $a_0 = \frac{2}{\sqrt{3}}r_0$ , wherein  $r_0 = l\sqrt{N}$  is denoted as the end-to-end distance of an unstretched chain. In current deformed state it reads:  $r = r_0\sqrt{3^{-1}\sqrt{\lambda_1^2 + \lambda_2^2 + \lambda_3^2}}$ , where  $\lambda_1, \lambda_2, \lambda_3$  are the principal stretches of the right Cauchy-Green tensor  $\mathbf{C} = \mathbf{F}^T \mathbf{F}$ ,  $\mathbf{F}$  being the deformation gradient. By defining the mean value of the stretch  $\lambda_c$ , which is required to determine the expression for the macroscopic free energy, we obtain

$$\lambda_c = \frac{r}{r_0} = \sqrt{\frac{1}{3} [\lambda_1^2 + \lambda_2^2 + \lambda_3^2]} = \sqrt{\frac{I_1}{3}}. \quad (4)$$

1 In Eqn (4),  $I_1$  is the first invariant of  $C$ , i.e.  $I_1 = \text{tr}C$ . Inserting the Langevin statistics of a single chain, the  
 2 free energy can be obtained as

$$\Psi_{8ch} = \mu N \left[ \gamma \lambda_{c,r} + \ln \left( \frac{\gamma}{\sinh \gamma} \right) \right], \quad (5)$$

3 with the relative chain stretch  $\lambda_{c,r} = \frac{\lambda_c}{\sqrt{N}} = \sqrt{\frac{I_1}{3N}}$ , where  $\mu$ ,  $N$ ,  $\gamma$  are the shear modulus, the number of chain  
 4 segments per chain and the inverse Langevin function, respectively. It is revealed that this model accurately  
 5 captures the ultimate strain of network deformation while requiring only two material parameters, the shear  
 6 modulus ( $\mu$ ) and the number of segments per chain ( $N$ ). Due to the inherent micromechanical explanations in  
 7 defining these two material parameters, such constitutive framework is often referred a micromechanically-  
 8 motivated model.



**Fig. 2** Performance of the eight-chain model on the Treloar data. The fittings and simulations in two deformation modes (UN and EB) are good to excellent except in the pure shear case where the prediction fails to reproduce the S-shape of uniaxial case at high strains. Uniaxial simulation and fitting are almost coincident (**left**) while equibiaxial simulation overestimates the Treloar data.

1 The analytical  $P_i(\lambda_i)$ -relations for the three deformation modes UN, EB and PS, which are required to check  
 2 the performance of the model on the Treloar data, are obtained from Eqn (1) as

$$P_1^{UN} = \frac{\mu^{UN}}{3} \left[ \frac{3N^{UN} - \lambda_{cu}^2}{N^{UN} - \lambda_{cu}^2} \right] [\lambda - \lambda^{-2}], \quad \lambda_{cu} = \sqrt{\frac{1}{3} \left[ \lambda^2 + \frac{2}{\lambda} \right]} \quad (6)$$

$$P_{1,2}^{EB} = \frac{\mu^{EB}}{3} \left[ \frac{3N^{EB} - \lambda_{cb}^2}{N^{EB} - \lambda_{cb}^2} \right] [\lambda - \lambda^{-5}], \quad \lambda_{cb} = \sqrt{\frac{1}{3} \left[ 2\lambda^2 + \frac{1}{\lambda^4} \right]} \quad (7)$$

$$P_1^{PS} = \frac{\mu^{PS}}{3} \left[ \frac{3N^{PS} - \lambda_{cp}^2}{N^{PS} - \lambda_{cp}^2} \right] [\lambda - \lambda^{-3}], \quad \lambda_{cp} = \sqrt{\frac{1}{3} [\lambda^2 + \lambda^{-2} + 1]}. \quad (8)$$

3 Note that in approximating the inverse Langevin function ( $\gamma$ ), the Padé approximation is applied due to its  
 4 superior performance over other approximation procedures, i.e.  $\gamma = \mathcal{L}^{-1}(\lambda_{c,r}) \approx \lambda_{c,r} \frac{3-\lambda_{c,r}^2}{1-\lambda_{c,r}^2}$ , cf. [8]. By  
 5 calibrating the UN, EB and PS equations to the corresponding Treloar data, the optimal material parameters  
 6 are estimated:

$$\mu^{UN} = 0.264 \text{ MPa}, \quad \mu^{EB} = 0.358 \text{ MPa}, \quad \mu^{PS} = 0.311 \text{ MPa},$$

$$N^{UN} = 25.60, \quad N^{EB} = 30.26, \quad N^{PS} = 51.32.$$

8 The optimal material parameters obtained by using analytical expressions are utilized to simulate the exper-  
 9 imental data of the other two deformation modes and their corresponding errors with respect to the Treloar  
 10 data are calculated. The results, cf. Fig (2) are obtained by inserting the Padé approximation of the inverse  
 11 Langevin function in the free energy function. It is clear from the Fig (2) that with a least number of material  
 12 parameters, i.e. two, this model produces better results in all deformation modes.

### 13 3.2 Modified Flory-Erman model

14 In most of the micromechanically-inspired models, authors derive the energy functions by considering only  
 15 the contributions from the cross-linking of polymer chains while the contributions due to entanglements have  
 16 been neglected. Boyce and Arruda [9] proposed a modification of their original model by adding a constraint  
 17 term devised by Flory and Erman [21]. The assumption of Flory and Erman is that a long macromolecule  
 18 network consists of numerous chain connection points, which are constrained from phantom characteristics  
 19 due to the presence of neighbouring chains. As a result, the elastic strain energy of the network is originated  
 20 from two contributions, i.e. the phantom ( $\Psi_{ph}$ ) and the topological constraint ( $\Psi_{ct}$ ) contributions as

$$\Psi = \Psi_{ph} + \Psi_{ct}. \quad (9)$$

21 Flory and Erman [21] derived the phantom part of the energy function from the Gaussian chain statistics,  
 22 equivalent to the Neo-Hookean case, i.e.  $\Psi_{ph} = \sum_{i=1}^3 \frac{\mu}{2} [1 - \frac{1}{\phi}] [\lambda_i^2 - 1]$  and the constraint part is obtained

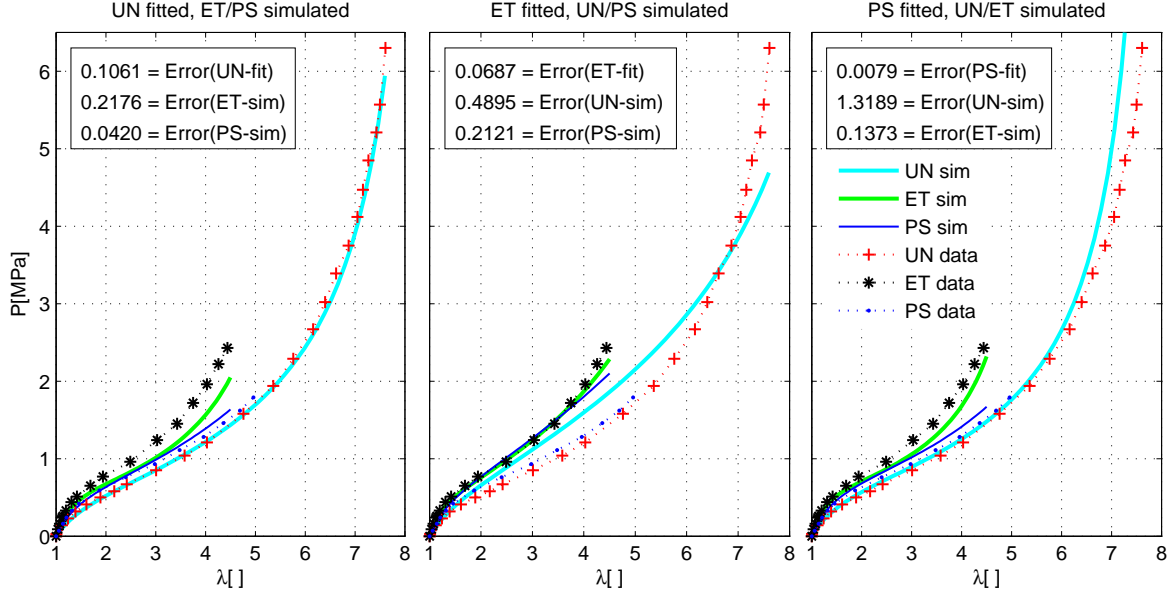
1 from the micromechanics of the chain molecules as

$$\Psi_{ct} = \sum_{i=1}^3 \frac{\mu}{2} \left[ B_i + D_i - \ln(B_i + 1) - \ln(D_i + 1) \right] \quad (10)$$

2 with  $B_i = \kappa^2 [\lambda_i^2 - 1] [\lambda_i^2 + \kappa]^{-2}$ ,  $D_i = \lambda_i^2 \kappa^{-1} B_i$ . In Eqn (10), the parameter  $\mu$  represents the shear modulus,  $\phi$   
 3 counts the number of chains joining at a junction and  $\kappa$  measures the strengths of the constraints, respectively.  
 4 Since the Flory-Erman model at large stretches deviates significantly from the actual response of the networks  
 5 due to the Gaussian nature of its phantom part, Boyce and Arruda [11] applied their eight-chain energy  
 6 function for the phantom part to improve the overall response, which, we designate here as the modified  
 7 Flory-Erman model. Replacing the Neo-Hookean part by the eight-chain energy function, the modified Flory-  
 8 Erman model yields

$$\begin{aligned} \Psi &= \Psi_{8ch} + \Psi_{ct} \\ &= \mu N \left[ \gamma \lambda_{c,r} + \ln \left( \frac{\gamma}{\sinh \gamma} \right) \right] + \sum_{i=1}^3 \frac{\mu}{2} \left[ B_i + D_i - \ln(B_i + 1) - \ln(D_i + 1) \right] \end{aligned} \quad (11)$$

9 where  $\mu, N, \gamma, \lambda_{c,r}$  are defined in Section 3.1.



**Fig. 3** Performance of the Flory-Erman model modified by the eight-chain energy function on the Treloar data. The fitting quality and predictions are excellent, especially if compared to the similarly structured models, e.g. the original eight-chain model or the Gent model, cf. Steinmann et al. [28].

10 The constraint contribution of the stress-stretch relations has to be derived from the single differentiation of  
 11 the constraint part of the energy function appearing in Eqn (11) and is demonstrated in Appendix A. If the



1 analytical expressions for UN, EB and PS fit to the corresponding Treloar data, the optimal material parameter  
2 sets are found to be

$$\begin{aligned} [\mu^{UN}, N^{UN}, \kappa^{UN}] &= [0.251 \text{ MPa}, 25.46, 2.0] \\ [\mu^{EB}, N^{EB}, \kappa^{EB}] &= [0.363 \text{ MPa}, 37.75, 0.011] \\ [\mu^{PS}, N^{PS}, \kappa^{PS}] &= [0.24 \text{ MPa}, 22.07, 5.323]. \end{aligned}$$

3 Each of the parameter sets is used to simulate the experimental data of other two deformation modes. The  
4 results are plotted in Fig (3) as well as the corresponding errors from both fitting and simulation of experiments  
5 not used for the parameter identification. In comparison to the original eight-chain model, it is interesting to  
6 note that the inclusion of the constraint contribution to the eight-chain energy function improves the simulation  
7 for the EB in all three deformation cases as well as the simulations for UN and EB by the PS-fitted parameters,  
8 cf. Fig 3 (**right**). A small drawback of the modified model is, the additional contribution for the topological  
9 constraint yields a complicated lengthy expression for the stress-stretch relationships, see Appendix A.

### 10 3.3 Gornet-Desmorat (GD) model

11 To improve the prediction capability in multi-axial loadings for rubber-like materials, very recently Gornet et  
12 al. [30] proposed a strain energy function based on the first and second strain invariants of the right Cauchy-  
13 Green tensor. The first part is dependent on the first invariant and it corresponds to the Hart-Smith model. The  
14 equivalence between the eight-chain model and  $I_1$ -dependent part of the Hart-Smith model is established by  
15 Chagnon et al. [19]. To enhance the phantom network, i.e. the cross-link part expressed by the first invariant  
16  $I_1$ , an additional function based on  $I_2$  is added to the energy function. Gornet et al. [30] show that such  
17 addition will constrain the eight-chains by a new network of chains on the surfaces of the cube, which mimics  
18 the behaviour of the constraint contribution. Finally, the energy function of the GD model is expressed as

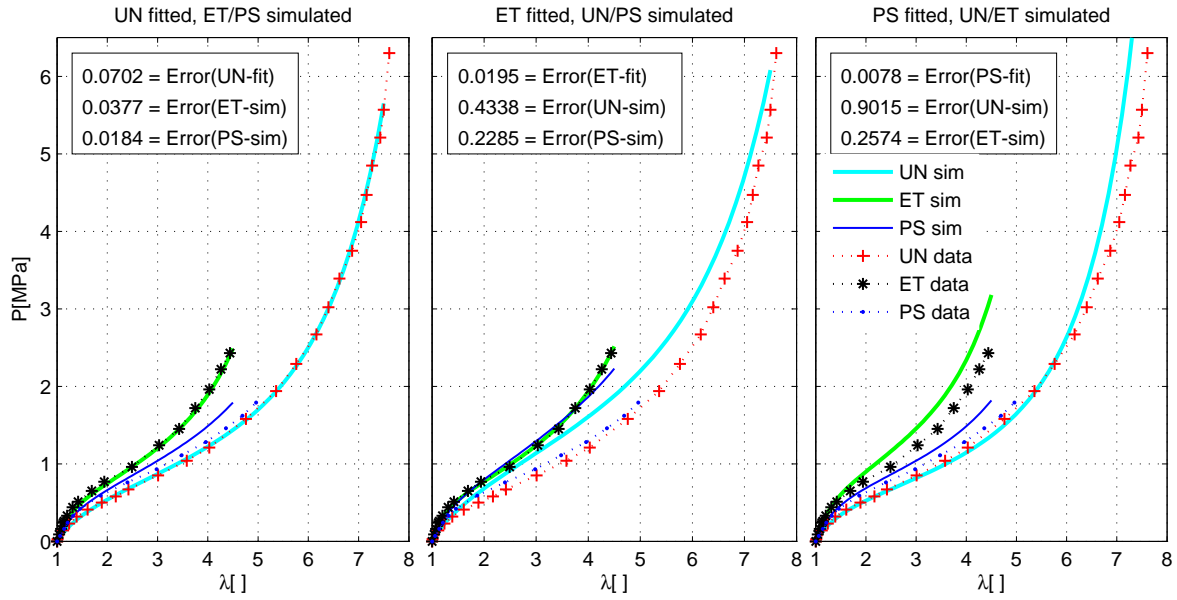
$$\Psi = h_1 \int e^{h_3[I_1-3]^2} dI_1 + 3h_2 \int \frac{1}{\sqrt{I_2}} dI_2 \quad (12)$$

19 where  $h_1, h_2, h_3$  are material parameters. Evaluation of Eqn (2) provides the corresponding analytical formu-  
20 lations for the UN, EB and PS cases:

$$P_1^{UN} = 2 \left[ h_1 e^{h_3 [I_1 - 3]^2} + \frac{3h_2}{\lambda \sqrt{I_2}} \right] \left[ \lambda - \lambda^{-2} \right] \quad (13)$$

$$P_{1,2}^{EB} = 2 \left[ h_1 e^{h_3 [I_1 - 3]^2} + \frac{3h_2 \lambda^2}{\sqrt{I_2}} \right] \left[ \lambda - \lambda^{-5} \right] \quad (14)$$

$$P_1^{PS} = 2 \left[ h_1 e^{h_3 [I_1 - 3]^2} + \frac{3h_2}{\sqrt{I_2}} \right] \left[ \lambda - \lambda^{-3} \right]. \quad (15)$$



**Fig. 4** Comparison between the GD model and the Treloar data. Fitting to the analytical formulations and model prediction quality are excellent, especially in UN and PS. Only predictions for the EB-fitted parameters overestimate the UN and PS data.

- 1 By calibrating the UN, EB and PS equations to the corresponding Treloar data, the optimal material parameters are found:

$$\begin{aligned} h_1^{UN} &= 0.142 \text{ MPa}, & h_1^{EB} &= 0.194 \text{ MPa}, & h_1^{PS} &= 0.124 \text{ MPa}, \\ h_2^{UN} &= 1.58\text{e-}2 \text{ MPa}, & h_2^{EB} &= 2.0\text{e-}14 \text{ MPa}, & h_2^{PS} &= 0.034 \text{ MPa}, \\ h_3^{UN} &= 3.49\text{e-}4, & h_3^{EB} &= 2.56\text{e-}4, & h_3^{PS} &= 4.94\text{e-}4. \end{aligned}$$

- 4 For validity of this model, each parameter set obtained from the optimization tool is used to simulate the other two deformation modes. The results are plotted in Fig (4) together with fitting and simulation errors in each case. It can clearly be shown here that the simulations for the EB and PS by the UN-fitted parameters produce results that are quantitatively much more acceptable than any other of the previous models with the same number of parameters, cf. Figs (4, **left**) and (4, **right**). The simulations for the EB-fitted parameters

1 overestimate the UN and PS data, cf. Fig (4, **middle**) while the predictions for the PS-fitted parameters are  
 2 good to fair, cf. Fig (4, **right**).

### 3 3.4 Bootstrapped eight-chain model

4 In an attempt to improve the results for the eight-chain model at small strain, a new model called bootstrapped  
 5 eight-chain model with only two material parameters (same as in the original eight-chain model) is proposed  
 6 by Miroshnychenko and Green [31; 32]. They further assume that, in order to improve results especially in the  
 7 biaxial deformation, an additive form of the strain energy function that involves two functions-one depends  
 8 on the first invariant and the other on the second invariant of a deformation tensor, has to be formulated. Such  
 9 an assertion is also supported by Pucci and Saccomandi [25], Hart-Smith [20]. The full strain energy function  
 10 of the bootstrapped eight-chain model is written as:

$$\Psi = \Psi_{8ch} \left( \frac{i_1}{\sqrt{3N}} - \frac{\lambda_c}{\sqrt{N}} \right) + \Psi_{8ch} \left( \frac{\lambda_c}{\sqrt{N}} \right) \quad (16)$$

11 where  $i_1 = \lambda_1 + \lambda_2 + \lambda_3$  and the definitions of  $\lambda_c, N$  are given in Section 3.1. For the complete expression  
 12 of the eight-chain model, previous subsection is referred. The energy function from Eqn (16) yields three  
 13 principal stresses as

$$\sigma_i = \mu\sqrt{N} \left[ \frac{\lambda_i}{\sqrt{3}} - \frac{\lambda_i^2}{3\lambda_c} \right] \mathcal{L}^{-1} \left( \frac{i_1}{\sqrt{3N}} - \frac{\lambda_c}{\sqrt{N}} \right) + \mu\sqrt{N} \left[ \frac{\lambda_i^2}{3\lambda_c} \right] \mathcal{L}^{-1} \left( \frac{\lambda_c}{\sqrt{N}} \right). \quad (17)$$

14 Using Eqn (17), the analytical formulations for the three different deformation modes, i.e. UN, EB and PS are  
 15 derived as

$$P_1^{UN} = \frac{\mu^{UN}}{\lambda} \lambda_u \frac{3N^{UN} - \lambda_u^2}{N^{UN} - \lambda_u^2} \left[ \frac{\lambda - \lambda^{-1/2}}{\sqrt{3}} - \frac{\lambda^2 - \lambda^{-1}}{3\lambda_{cu}} \right] + \frac{\mu^{UN}}{3} \left[ \frac{3N^{UN} - \lambda_{cu}^2}{N^{UN} - \lambda_{cu}^2} \right] \left[ \lambda - \lambda^{-2} \right] \quad (18)$$

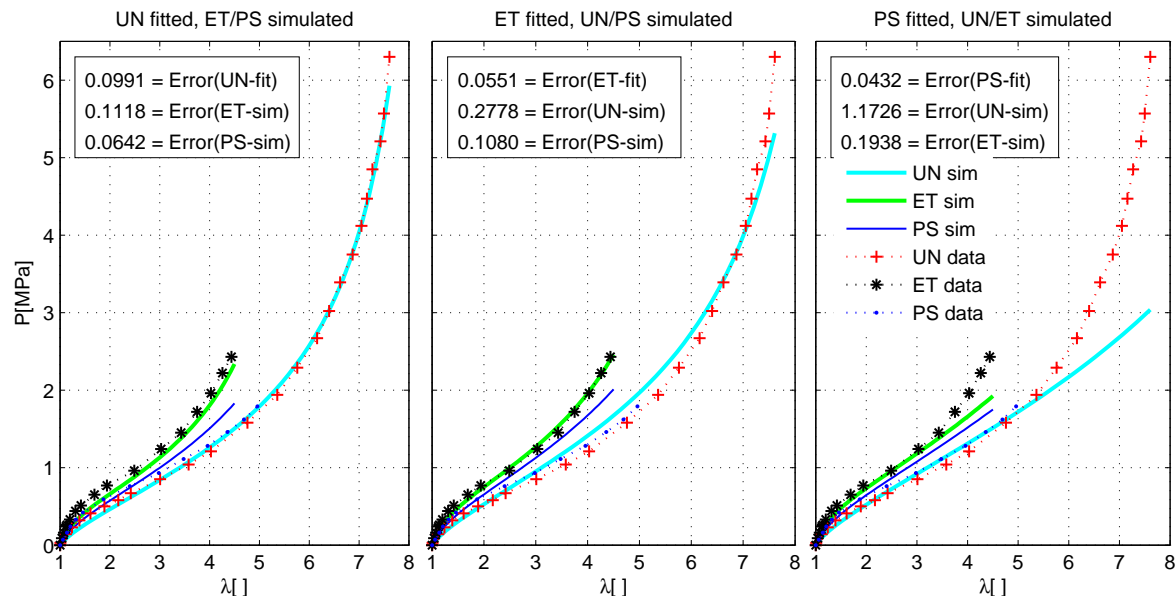
$$P_{1,2}^{EB} = \frac{\mu^{EB}}{\lambda} \lambda_b \frac{3N^{EB} - \lambda_b^2}{N^{EB} - \lambda_b^2} \left[ \frac{\lambda - \lambda^{-2}}{\sqrt{3}} - \frac{\lambda^2 - \lambda^{-4}}{3\lambda_{cb}} \right] + \frac{\mu^{EB}}{3} \left[ \frac{3N^{EB} - \lambda_{cb}^2}{N^{EB} - \lambda_{cb}^2} \right] \left[ \lambda - \lambda^{-5} \right] \quad (19)$$

$$P_1^{PS} = \frac{\mu^{PS}}{\lambda} \lambda_p \frac{3N^{PS} - \lambda_p^2}{N^{PS} - \lambda_p^2} \left[ \frac{\lambda - \lambda^{-1}}{\sqrt{3}} - \frac{\lambda^2 - \lambda^{-2}}{3\lambda_{cp}} \right] + \frac{\mu^{PS}}{3} \left[ \frac{3N^{PS} - \lambda_{cp}^2}{N^{PS} - \lambda_{cp}^2} \right] \left[ \lambda - \lambda^{-3} \right]. \quad (20)$$

16 In Eqns (18-20),  $\lambda_u = \frac{i_{1u}}{\sqrt{3}} - \lambda_{cu}$ ,  $\lambda_b = \frac{i_{1b}}{\sqrt{3}} - \lambda_{cb}$ ,  $\lambda_p = \frac{i_{1p}}{\sqrt{3}} - \lambda_{cp}$ ,  $i_{1u} = \lambda + 2\lambda^{-1/2}$ ,  $i_{1b} =$   
 17  $2\lambda + \lambda^{-2}$ ,  $i_{1p} = \lambda + \lambda^{-1} + 1$  and  $\lambda_{cu}, \lambda_{cb}, \lambda_{cp}$  are defined in Eqns (6-8).

18 Fitting the analytical equations (18-20) to Treloar's data yields the following parameter sets

$$\begin{aligned} \mu^{UN} &= 0.277 \text{ MPa}, & \mu^{EB} &= 0.318 \text{ MPa}, & \mu^{PS} &= 0.322 \text{ MPa}, \\ N^{UN} &= 26.50, & N^{EB} &= 30.11, & N^{PS} &= 72.18. \end{aligned}$$



**Fig. 5** Performance of the Bootstrapped eight-chain model on the Treloar data. The fittings and simulations are good to excellent, especially if compared to the similarly structured models, e.g. the original eight-chain model or the two-parameter Gent model.

1 Figure (5) depicts all fits, the simulations of complementary deformation modes that are not used during  
 2 optimization and the deviations in comparison to the Treloar's data. Both fitting quality and validity of the  
 3 identified parameters are very good to excellent in the UN and EB-cases. Optimization with respect to the  
 4 PS-data provides parameter sets that fail to predict the UN-data, cf. Fig (5, **right**). Similar to the classical  
 5 eight-chain model, the bootstrapped model also fails to predict the S-shape in the case of UN for PS-fitted  
 6 parameters. After analysing the errors, fittings and the validation results, it can be concluded that with the same  
 7 number of parameters, the bootstrapped eight-chain model illustrates better performance in all deformation  
 8 modes than the classical eight-chain model.

### 9 3.5 Meissner-Matějka model

10 Based on the fact that to cope with the experimental data, not only the contribution from the cross-link part but  
 11 also a constraint term in the energy function is essential. Meissner and Matějka [34; 35] proposed an energy  
 12 function where the cross-link term is derived from the structure-based Arruda-Boyce model and the constraint  
 13 part is based on the first invariant of the generalized deformation tensor. Note that here the constraint part is  
 14 exactly same as the constraint part in the extended tube model proposed by Kalikse and Heinrich [18]. Then,  
 15 the total energy function is expressed:

$$\Psi = \mu N \left[ \gamma \lambda_{c,r} + \ln \left( \frac{\gamma}{\sinh \gamma} \right) \right] + \sum_{i=1}^3 \frac{2\mu_e}{\beta^2} \left[ \lambda_i^{-\beta} - 1 \right] \quad (21)$$

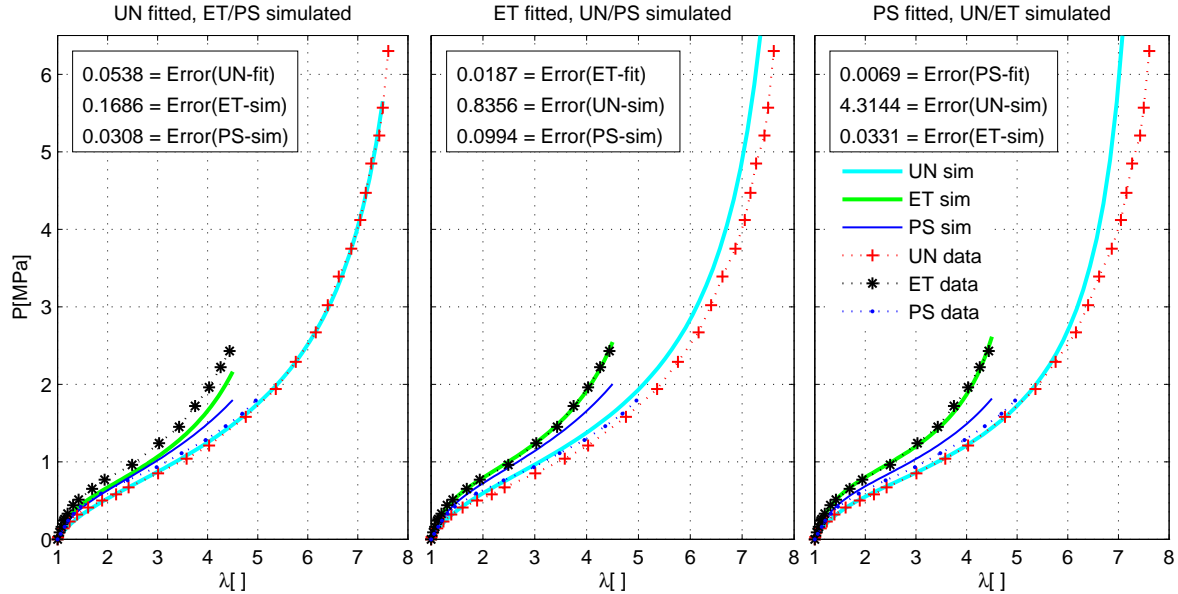
- 1 where  $\mu, N, \mu_e, \beta$  ( $0 < \beta \leq 1$ ) are material parameters. The definitions of  $\lambda_{c,r}, \gamma$  are given in Section 3.1.
- 2 Evaluation of Eqn (1) provides the corresponding analytical formulations for the UN, EB and PS cases:

$$P_1^{UN} = \frac{\mu^{UN}}{3} \left[ \frac{3N^{UN} - \lambda_{cu}^2}{N^{UN} - \lambda_{cu}^2} \right] \left[ \lambda - \lambda^{-2} \right] + \frac{2\mu_e^{UN}}{\beta^{UN}} \left[ \lambda^{\frac{\beta^{UN}}{2}-1} - \lambda^{-\beta^{UN}-1} \right] \quad (22)$$

$$P_{1,2}^{EB} = \frac{\mu^{EB}}{3} \left[ \frac{3N^{EB} - \lambda_{cb}^2}{N^{EB} - \lambda_{cb}^2} \right] \left[ \lambda - \lambda^{-5} \right] + \frac{2\mu_e^{EB}}{\beta^{EB}} \left[ \lambda^{2\beta^{EB}-1} - \lambda^{-\beta^{EB}-1} \right] \quad (23)$$

$$P_1^{PS} = \frac{\mu^{PS}}{3} \left[ \frac{3N^{PS} - \lambda_{cp}^2}{N^{PS} - \lambda_{cp}^2} \right] \left[ \lambda - \lambda^{-3} \right] + \frac{2\mu_e^{PS}}{\beta^{PS}} \left[ \lambda^{\beta^{PS}-1} - \lambda^{-\beta^{PS}-1} \right] \quad (24)$$

- 3 where  $\lambda_{cu}, \lambda_{cb}, \lambda_{cp}$  are defined in Eqns (6-8). If one fits the UN, EB and PS equations to the corresponding
- 4 Treloar data, the optimal material parameters are found to be:



**Fig. 6** Performance of the Meissner-Matějka model on the Treloar data. The fitting quality and model predictions are excellent in all deformation modes.

$$[\mu^{UN}, \mu_e^{UN}, N^{UN}] = [0.256 \text{ MPa}, 0.055 \text{ MPa}, 25.37]$$

$$[\mu^{EB}, \mu_e^{EB}, N^{EB}] = [0.270 \text{ MPa}, 0.095 \text{ MPa}, 23.47]$$

$$[\mu^{PS}, \mu_e^{PS}, N^{PS}] = [0.217 \text{ MPa}, 0.146 \text{ MPa}, 20.37].$$

1 Concerning validity of this model, all resulting curves and corresponding errors in comparison with Treloar's  
 2 data are summarized in Fig (6). It reveals excellent fitting and validity of the model in all deformation modes.  
 3 During optimization for the parameter identification, an important parameter, i.e.  $\beta$  ( $0 < \beta \leq 1$ ) is kept frozen  
 4 ( $\beta = 0.2$ ) as suggested by Kaliske and Heinrich [17] for better fitting and simulation of the Treloar data,  
 5 even if it can be determined during parameter identification. Note that due to the presence of the constraint  
 6 contribution in the energy function, the simulations for the equibiaxial case are better than the original eight-  
 7 chain model. The predictions for the UN and PS cases by the EB-fitted parameters slightly overestimate the  
 8 Treloar data, cf. Fig (6, **middle**). In addition, it can be noted that this model captures all deformation mode data  
 9 excellently if it is compared to the similarly structured 3-parameters model, e.g. the modified Flory-Erman  
 10 model or the Gornet-Desmorat (GD) model.

### 11 3.6 Bechir model

12 As mentioned earlier, the classical eight-chain model ignores the contribution from the entanglements-like  
 13 physical cross-links during deformation of the network. To add the effects of the interactions between chains  
 14 of the cross-linked network, an extra energy function, in addition to the eight-chain energy function, is ap-  
 15 pended. The free-energy of the constraint network is idealized using the standard three-chain energy function  
 16 and the free-energy of the unconstrained idealized network is constructed by means of the eight-chain model.  
 17 Therefore, according to Bechir et al. [33], the total free energy function of this model is merely a combination  
 18 of the two chain models, i.e.

$$\begin{aligned} \Psi &= \Psi_{8ch} + \Psi_{3ch} \\ &= \mu_f N_8 \left[ \lambda_{c,r} \beta + \ln \left( \frac{\beta}{\sinh \beta} \right) \right] + \frac{\mu_c N_3}{3} \sum_{j=1}^{j=3} \left[ \bar{\lambda}_{rj} \bar{\beta}_j + \ln \left( \frac{\bar{\beta}_j}{\sinh \bar{\beta}_j} \right) \right] \end{aligned} \quad (25)$$

19 where  $\beta = \mathcal{L}^{-1}(\lambda_{c,r})$ ,  $\lambda_{c,r} = \sqrt{\frac{\lambda_1^2 + \lambda_2^2 + \lambda_3^2}{3N_8}}$  and  $\bar{\beta}_j = \mathcal{L}^{-1}(\bar{\lambda}_{rj})$ ,  $\bar{\lambda}_{rj} = \frac{k\lambda_j}{\sqrt{N_3}}$ . In Eqn (25),  $\mu_f$ ,  $\mu_c$ ,  $N_8$ ,  $N_3$ ,  $k$   
 20 are shear moduli for the free and constraint parts, numbers of chain segments for the eight and three-chain  
 21 functions and a non-affine parameter, respectively. Bechir et al. [33] also proposed some relations for the ma-  
 22 terial parameter set, e.g.,  $\mu_o = \mu_c + \mu_f$ , where  $\mu_c = [1 - \frac{\eta\alpha}{\sqrt{N_3}}]\mu_o$ ,  $\mu_f = \rho\lambda_r\mu_o$ , and  $\alpha = \max(\bar{\lambda}_1, \bar{\lambda}_2, \bar{\lambda}_3)$ . Note  
 23 that  $\bar{\lambda}_i$  relate to the principal micro-stretches while  $\lambda_i$  are the line-stretches (continuum stretches). Evaluation  
 24 of Eqn (1) provides the corresponding analytical formulations for the UN, EB and PS cases:

$$P_1^{UN} = \left[ 1 - \frac{\eta^{UN} \lambda_k}{\sqrt{N_3^{UN}}} \right] \frac{\mu^{UN}}{3\lambda} \left[ \lambda_k^2 \frac{3N_3^{UN} - \lambda_k^2}{N_3^{UN} - \lambda_k^2} - \lambda_k^{-1} \frac{3N_3^{UN} - \lambda_k^{-1}}{N_3^{UN} - \lambda_k^{-1}} \right] + \frac{\rho^{UN} \mu^{UN} \lambda_{c,ru}}{3} \left[ \frac{3 - \lambda_{ru}^2}{1 - \lambda_{ru}^2} \right] \left[ \lambda - \lambda^{-2} \right] \quad (26)$$

$$P_{1,2}^{EB} = \left[ 1 - \frac{\eta^{EB} \lambda_k}{\sqrt{N_3^{EB}}} \right] \frac{\mu^{EB}}{3\lambda} \left[ \lambda_k^2 \frac{3N_3^{EB} - \lambda_k^2}{N_3^{EB} - \lambda_k^2} - \lambda_k^{-4} \frac{3N_3^{EB} - \lambda_k^{-4}}{N_3^{EB} - \lambda_k^{-4}} \right] + \frac{\rho^{EB} \mu^{EB} \lambda_{c,rb}}{3} \left[ \frac{3 - \lambda_{rb}^2}{1 - \lambda_{rb}^2} \right] \left[ \lambda - \lambda^{-5} \right] \quad (27)$$

$$P_1^{PS} = \left[ 1 - \frac{\eta^{PS} \lambda_k}{\sqrt{N_3^{PS}}} \right] \frac{\mu^{PS}}{3\lambda} \left[ \lambda_k^2 \frac{3N_3^{PS} - \lambda_k^2}{N_3^{PS} - \lambda_k^2} - \lambda_k^{-2} \frac{3N_3^{PS} - \lambda_k^{-2}}{N_3^{PS} - \lambda_k^{-2}} \right] + \frac{\rho^{PS} \mu^{PS} \lambda_{c,rp}}{3} \left[ \frac{3 - \lambda_{rp}^2}{1 - \lambda_{rp}^2} \right] \left[ \lambda - \lambda^{-3} \right] \quad (28)$$

where  $\lambda_k = k\lambda$ ,  $\lambda_{c,ru} = \sqrt{\frac{\lambda^2 + 2/\lambda}{3N_8}}$ ,  $\lambda_{c,rb} = \sqrt{\frac{2\lambda^2 + 1/\lambda^4}{3N_8}}$ ,  $\lambda_{c,rp} = \sqrt{\frac{\lambda^2 + 1 + 1/\lambda^2}{3N_8}}$ . Fitting UN, EB and

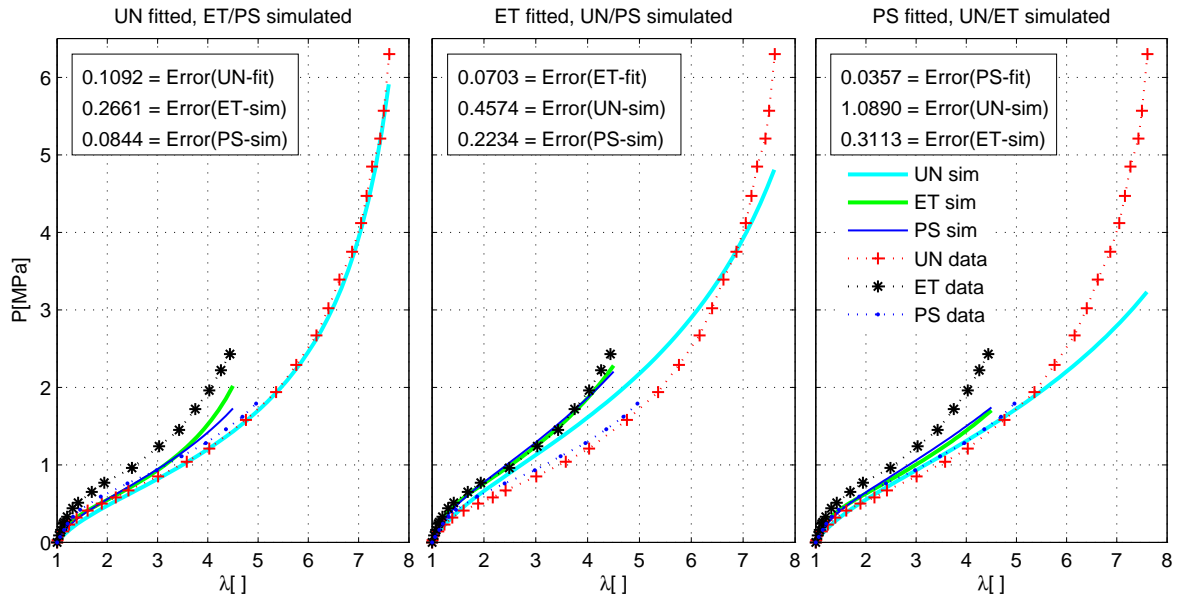


Fig. 7 Performance of the Bechir model on the Treloar data.

$$\begin{aligned}
[\mu^{UN}, \rho^{UN}, \eta^{UN}, N_3^{UN}, N_8^{UN}] &= [0.296 \text{ MPa}, 1.28, 1.39, 71.83, 26.01] \\
[\mu^{EB}, \rho^{EB}, \eta^{EB}, N_3^{EB}, N_8^{EB}] &= [0.251 \text{ MPa}, 1.46, 1.20, 68.20, 22.02] \\
[\mu^{PS}, \rho^{PS}, \eta^{PS}, N_3^{PS}, N_8^{PS}] &= [0.201 \text{ MPa}, 1.26, 1.02, 56.21, 20.21].
\end{aligned}$$

1 To study the model performance, each parameter set obtained from the optimization tool is used to simulate  
2 the other two deformation modes. The results are plotted in Fig (7) together with fitting and simulation errors  
3 in each case. Although, this model inherits more parameters, i.e. five in total, but it can be clearly seen that  
4 each set of optimal parameters produces simulation results for the complementary deformation modes that  
5 are not quantitatively improved than any other of the previous models of less parameters.

### 6 3.7 Wu-Giessen model

7 Several authors [6; 7; 14; 24] coin the term *full-network* where the chains are assumed to be randomly oriented  
8 in space for which the strain energy function is derived by integrating the response of all chains over the space.  
9 Since the numerical integration for such a full-network model is computationally costly, a weighted average  
10 is proposed by combining the three-chain and the eight-chain formulations which might provide better results  
11 than the individual three-chain or the eight-chain model [14], i.e.

$$\Psi = \Psi_{3c}[1 - \rho] + \rho\Psi_{8c} \quad (29)$$

12 where the parameter  $\rho$  is a constant or related to some other physical quantity which is for instance related  
13 to the deformation process and  $\Psi_{3c}, \Psi_{8c}$  are energy functions for the three-chain and eight-chain models,  
14 respectively. Such form of the full-network model was proposed by Wu and Giessen [14] to improve the mod-  
15 elling capacity for amorphous glassy polymers, e.g. polycarbonate, where the hyperelastic energy function  
16 is used for modelling the so-called back stress. Frequently used relation for  $\rho$  is  $\rho = 0.85\lambda_{\max}/\sqrt{N}$ , where  
17  $\lambda_{\max} = \max(\lambda_1, \lambda_2, \lambda_3)$ . The factor 0.85 is chosen to provide the best correlation of Eqn (29) with numer-  
18 ical integration of the full-network equation. The analytical expressions for UN, EB and PS are obtained,  
19 according to the idea of the Wu and Giessen [14] model, as

$$P_1^{UN} = P_{1,3c}^{UN}[1 - \rho] + \rho P_{1,8c}^{UN}, \quad P_1^{EB} = P_{1,3c}^{EB}[1 - \rho] + \rho P_{1,8c}^{EB}, \quad P_1^{PS} = P_{1,3c}^{PS}[1 - \rho] + \rho P_{1,8c}^{PS} \quad (30)$$



1 where  $P_{1,8c}^{UN}, P_{1,8c}^{EB}, P_{1,8c}^{PS}$  and  $P_{1,3c}^{UN}, P_{1,3c}^{EB}, P_{1,3c}^{PS}$  are UN, EB, PS stresses for the eight-chain and three-chain  
 2 models, respectively. For the derivations of  $P_{1,3c}^{UN}, P_{1,3c}^{EB}$ , and  $P_{1,3c}^{PS}$ , our previous work is referred [28]. To  
 3 verify the sensitivity of the model with respect to different deformation modes and also with material param-  
 4 eters, if the UN, EB and PS equations fit to the corresponding Treloar data, the optimal material parameters  
 5 are identified:

$$6 \quad \begin{aligned} \mu^{UN} &= 0.318 \text{ MPa}, & \mu^{EB} &= 0.403 \text{ MPa}, & \mu^{PS} &= 0.309 \text{ MPa}, \\ N^{UN} &= 63.69, & N^{EB} &= 58.00, & N^{PS} &= 90.53. \end{aligned}$$

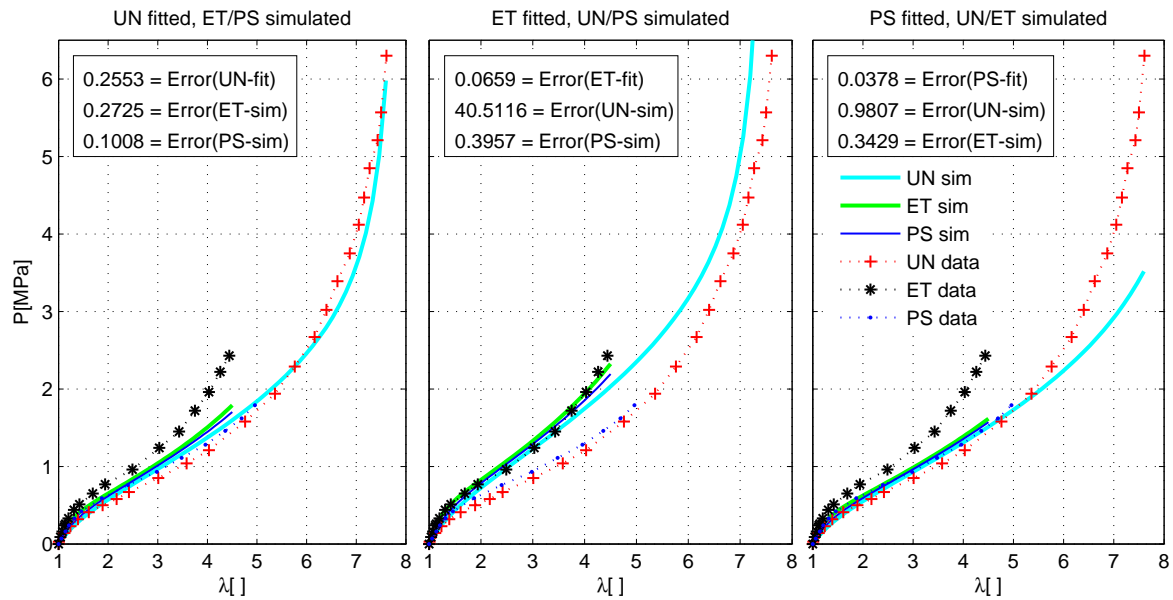


Fig. 8 Performance of the full-network model on the Treloar data.

7 Similar to other models, each parameter set of this model obtained from the fitting procedure is used to  
 8 simulate the other two deformation modes and the corresponding errors are tabulated. The full-network model  
 9 suggested by Wu and Giessen [14] predicts a biaxial stress-stretch response which falls in-between the results  
 10 predicted by the eight-chain model and that predicted by the three-chain model. Aiming at better performance  
 11 over the eight-chain model or the three-chain model, the capability of this model to reproduce the experimental  
 12 data for all deformation modes does not show any significant improvement so far, cf. Fig (8).

### 13 3.8 Zuniga-Beatty model

14 Zuniga and Beatty [10] at first amend the original eight-chain and three-chain models by using a modified non-  
 15 Gaussian (Kuhn-Grün) probability function in the case of energy function derivation. Jernigan and Flory [22]

1 show that an amended distribution function, in contrast to the Kuhn-Grün function, provides a much improved  
 2 approximation to the exact result over the entire range of  $\lambda_r$  (relative chain stretch) values. Therefore, Zuniga  
 3 and Beatty [10] re-derive the energy functions for the eight-chain and three-chain models using the amended  
 4 distribution function. Then, a modified full-network model is proposed by combining the amended versions  
 5 of the eight-chain and three-chain models in the line of Wu and Giessen [14] concept. Wu and Giessen [14]  
 6 assume the same value for the number of chain segments, i.e.  $N$  for the eight-chain and three-chain models,  
 7 which is unjustified due to inherent geometrical considerations of these two models. Hence, Zuniga and Beatty  
 8 [10] propose separate values for  $N$ , i.e.  $N_3, N_8$  for the three-chain and eight-chain models, respectively. Due  
 9 to the amended form for the probability function, the energy functions of the eight-chain and three-chain  
 10 models are as follows

$$\Psi_{8c} = \mu \left[ N_8 \left[ \lambda_{c,r} \beta + \ln \left( \frac{\beta}{\sinh \beta} \right) \right] - \ln \left( \frac{\beta}{\lambda_r} \right) \right] \quad (31)$$

11 and

$$\Psi_{3c} = \frac{\mu}{3} \sum_{j=1}^{j=3} \left[ N_3 \left[ \lambda_{rj} \beta_j + \ln \left( \frac{\beta_j}{\sinh \beta_j} \right) \right] - \ln \left( \frac{\beta_j}{\lambda_{rj}} \right) \right]. \quad (32)$$

12 In Eqns (31) and (32),  $\lambda_{c,r} = \sqrt{\frac{I_1}{3N_8}}, \beta = \mathcal{L}^{-1}(\lambda_{c,r}), \beta_j = \mathcal{L}^{-1}(\lambda_{rj}), \lambda_{rj} = \frac{\lambda_j}{\sqrt{N_3}}$ , where  $I_1$  and  $\lambda_{c,r}$  are  
 13 defined in Section 3.1. Hence, the modified form of the full-network energy function proposed by Zuniga and  
 14 Beatty [10] is

$$\Psi = [1 - \rho_{3c}] \Psi_{3c} + \rho_{8c} \Psi_{8c} \quad (33)$$

15 where

$$\rho_{3c} = \frac{a \Lambda_L}{\sqrt{N_3}}, \quad \rho_{8c} = \frac{b \Lambda_c}{\sqrt{N_8}}. \quad (34)$$

16 In Eqn (34),  $\Lambda_L$  is related to the principal stretches through the state of deformation to which the continuum  
 17 is subjected so that the greater stretch  $\lambda_1 = \Lambda_L$  and the chain stretch for the eight-chain model is  $\Lambda_c =$   
 18  $\sqrt{\frac{\lambda_1^2 + \lambda_2^2 + \lambda_3^2}{3}}$  while  $a, b$  are two positive scaling factors. However, for simplicity, they choose to follow  $a =$   
 19  $b = 1$ , see Zuniga and Beatty [10] for details. The analytical formulations for the UN, EB and PS for this  
 20 composite model are

$$P_1^{UN} = P_{1,3cm}^{UN} \left[ 1 - \frac{\lambda}{\sqrt{N_3^{UN}}} \right] + \frac{\lambda_{cu}}{\sqrt{N_8^{UN}}} P_{1,8cm}^{UN} \quad (35)$$

$$P_1^{EB} = P_{1,3cm}^{EB} \left[ 1 - \frac{\lambda}{\sqrt{N_3^{EB}}} \right] + \frac{\lambda_{cb}}{\sqrt{N_8^{EB}}} P_{1,8cm}^{EB} \quad (36)$$

$$P_1^{PS} = P_{1,3cm}^{PS} \left[ 1 - \frac{\lambda}{\sqrt{N_3^{PS}}} \right] + \frac{\lambda_{cp}}{\sqrt{N_8^{PS}}} P_{1,8cm}^{PS} \quad (37)$$

1 where the expressions for  $P_{1,3cm}^{UN}$ ,  $P_{1,3cm}^{EB}$ ,  $P_{1,3cm}^{PS}$ ,  $P_{1,8cm}^{UN}$ ,  $P_{1,8cm}^{EB}$ ,  $P_{1,8cm}^{PS}$  can be obtained from Appendix B  
 2 and  $\lambda_{cu}$ ,  $\lambda_{cb}$ ,  $\lambda_{cp}$  are defined in Eqns (6-8). The optimal parameter sets for this model are estimated as

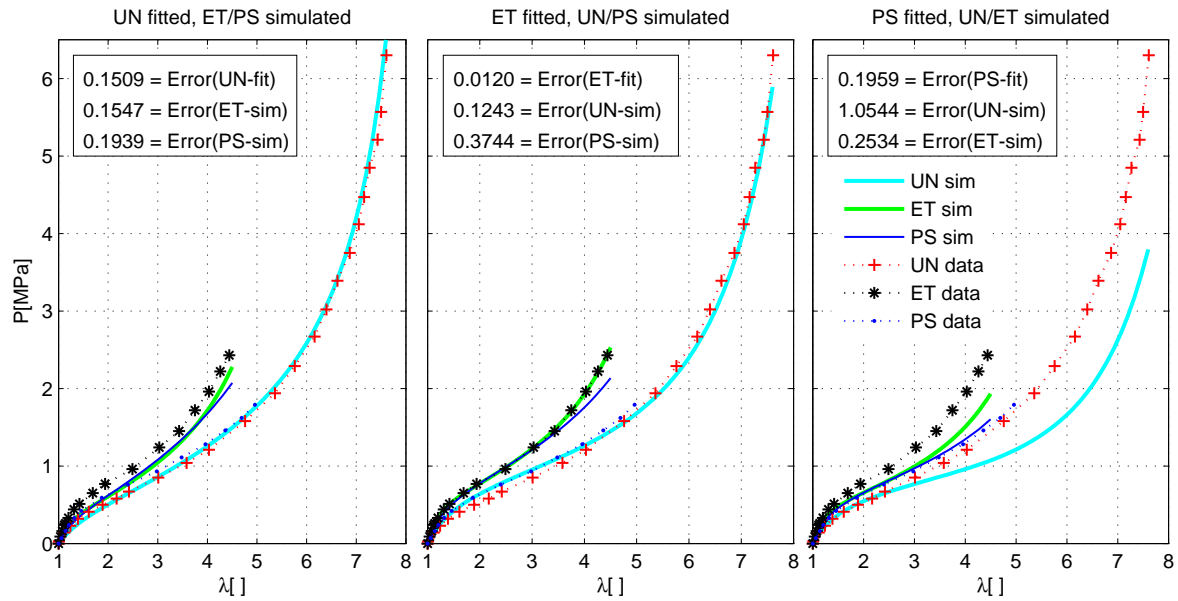


Fig. 9 Performance of the Zuniga-Beatty version of the full-network model on the Treloar data. The fitting and simulations in all deformation modes are excellent except an underestimation occurs in the case of UN data for the PS-fitted parameters.

$$\mu^{UN} = 0.27 \text{ MPa}, \quad \mu^{EB} = 0.48 \text{ MPa}, \quad \mu^{PS} = 0.45 \text{ MPa},$$

$$N_3^{UN} = 75.13, \quad N_3^{EB} = 8.8, \quad N_3^{PS} = 6.55,$$

$$N_8^{UN} = 25.12, \quad N_8^{EB} = 28.55, \quad N_8^{PS} = 31.49.$$

3  
 4 Concerning the validity of the model, the parameter set obtained from the optimization tool in each defor-  
 5 mation case is used to simulate the other two deformation modes. The results are plotted in Fig (9) together  
 6 with fitting and simulation errors in each case. The fittings and simulations for UN- and EB-fitted parameters  
 7 are good to excellent in predicting the data for other deformation modes except a small overestimation is  
 8 observed for the PS data. It can be clearly stated here that with an additional parameter, the Zuniga-Beatty

1 version of the full-network shows better performance than the Wu-Giessen version. The reason might be that  
 2 (a) Zuniga-Beatty used an improved version of the probability function and (b) the scaling factor ( $\rho$ , in the  
 3 original version of Wu-Giessen model) is replaced by a deformation-dependent stretch, cf. Eqn (34).

#### 4 **4 Conclusion**

5 In this paper, a comparative study on the classical eight-chain and full-network models and six of their mod-  
 6 ified versions is presented. Some of the modified versions are of phenomenological types while others are  
 7 originated from the micro-mechanics of chain molecules. In order to perform the comparative study, the ana-  
 8 lytical formulations in the case of three different deformation modes of the eight selected models are derived.  
 9 Then, the performance evaluation is highlighted of all selected models in reproducing the classical experimen-  
 10 tal data of Treloar. This study demonstrates that the two-parameter bootstrapped eight-chain model predicts  
 11 the Treloar data better than the classical eight-chain model. Note that several constitutive models, e.g. the  
 12 modified Flory-Erman model, the Meissner-Matějka model, the GD model which consider constraint contri-  
 13 bution in deriving their free energy functions predict all three deformation modes quite reasonable than the  
 14 classical eight-chain model. However, the Bechir model with five parameters does not show improved results  
 15 in comparison to the original eight-chain model. In the case of full-network model, the Zuniga-Beatty ver-  
 16 sion with an improved probability function for chain statistics shows better performance than the Wu-Giessen  
 17 form of full-network model. Such performance analysis will help a design engineer or a beginner in rubber  
 18 mechanics to choose whether the classical eight-chain model or full-network model or one of their modified  
 19 versions is appropriate from a considerable number of rubber-like material models available in the literature.

20

#### 21 **Appendix A: Derivatives of the Flory-Erman energy function (constraint part)**

22 To derive the analytical formulations for different deformation modes, the single derivatives of the energy  
 23 function (constraint part) of the Flory-Erman model expressed in principal stretches are essential, i.e.

$$\frac{\partial \Psi_{ct}}{\partial \lambda_i} = \frac{2\kappa^2[\lambda_i^2 - 1][\kappa^4 + \kappa^3\lambda_i^4 - \kappa^3\lambda_i^2 + 2\kappa^3 + 2\kappa^2\lambda_i^4 + 2\kappa\lambda_i^6 + \lambda_i^6]}{\lambda_i[[\lambda_i^2 + \kappa]^2][\kappa^4 + \kappa^3\lambda_i^4 + \kappa^3\lambda_i^2 + 2\kappa^3 + 3\kappa^2\lambda_i^4 + 3\kappa^2\lambda_i^2 + \kappa\lambda_i^6 + 3\kappa\lambda_i^4 + \lambda_i^6]}. \quad (38)$$

#### 24 **Appendix B: Analytical expressions for the amended three-chain and eight-chain models**

25 The analytical expressions for the amended three-chain and eight-chain models for different deformation  
 26 modes, i.e. UN, EB and PS are as follows,

27 Uniaxial tension (UN):

$$P_{1,3cm}^{UN} = \frac{\mu^{UN} \sqrt{N_3^{UN}}}{3} \left[ \beta_1 - \frac{\beta_2}{\lambda^{3/2}} + \frac{1}{\lambda^{3/2} N_3^{UN} \beta_2 \left[ 1 - \frac{1}{\lambda N_3^{UN}} - \frac{2}{\beta_2 \sqrt{\lambda N_3^{UN}}} \right]} - \frac{1}{N_3^{UN} \beta_1 \left[ 1 - \frac{\lambda^2}{N_3^{UN}} - \frac{2\lambda}{\beta_1 \sqrt{N_3^{UN}}} \right]} \right]$$

$$P_{1,8cm}^{UN} = \frac{\mu^{UN} [\lambda - \lambda^{-2}]}{3\lambda_{c,r}} \left[ \beta + \frac{1}{N_8^{UN} \lambda_{c,r}} - \frac{1}{N_8^{UN} \beta \left[ 1 - \lambda_{c,r}^2 - \frac{2\lambda_{c,r}}{\beta} \right]} \right]$$

$$\text{where, } \lambda_{c,r} = \sqrt{\frac{1}{3N_8^{UN}} \left[ \lambda^2 + \frac{2}{\lambda} \right]}, \beta = \lambda_{c,r} \frac{3 - \lambda_{c,r}^2}{1 - \lambda_{c,r}^2}, \beta_1 = \frac{\lambda}{\sqrt{N_3^{UN}}} \frac{3N_3^{UN} - \lambda^2}{N_3^{UN} - \lambda^2}, \beta_2 = \frac{1}{\sqrt{\lambda N_3^{UN}}} \frac{3N_3^{UN} \lambda - 1}{N_3^{UN} \lambda - 1}.$$

1 Equibiaxial tension (EB):

$$P_{1,3cm}^{EB} = \frac{\mu^{EB} \sqrt{N_3^{EB}}}{3} \left[ \beta_1 - \frac{\beta_3}{\lambda^3} + \frac{1}{N_3^{EB} \beta_3 \left[ \lambda^3 - \frac{1}{\lambda N_3^{EB}} - \frac{2\lambda}{\beta_3 \sqrt{N_3^{EB}}} \right]} - \frac{1}{N_3^{EB} \beta_1 \left[ 1 - \frac{\lambda^2}{N_3^{EB}} - \frac{2\lambda}{\beta_1 \sqrt{N_3^{EB}}} \right]} \right]$$

$$P_{1,8cm}^{EB} = \frac{\mu^{EB} [\lambda - \lambda^{-5}]}{3\lambda_{c,r}} \left[ \beta + \frac{1}{N_8^{EB} \lambda_{c,r}} - \frac{1}{N_8^{EB} \beta \left[ 1 - \lambda_{c,r}^2 - \frac{2\lambda_{c,r}}{\beta} \right]} \right]$$

$$\text{where, } \lambda_{c,r} = \sqrt{\frac{1}{3N_8^{EB}} \left[ 2\lambda^2 + \frac{1}{\lambda^4} \right]}, \beta = \lambda_{c,r} \frac{3 - \lambda_{c,r}^2}{1 - \lambda_{c,r}^2}, \beta_1 = \frac{\lambda}{\sqrt{N_3^{EB}}} \frac{3N_3^{EB} - \lambda^2}{N_3^{EB} - \lambda^2}, \beta_3 = \frac{\lambda^{-2}}{\sqrt{N_3^{EB}}} \frac{3N_3^{EB} - \lambda^{-4}}{N_3^{EB} - \lambda^{-4}}.$$

2 Pure shear (PS):

$$P_{1,3cm}^{PS} = \frac{\mu^{PS} \sqrt{N_3^{PS}}}{3} \left[ \beta_1 - \frac{\beta_4}{\lambda^2} + \frac{1}{N_3^{PS} \beta_4 \left[ \lambda^2 - \frac{1}{N_3^{PS}} - \frac{2\lambda}{\beta_4 \sqrt{N_3^{PS}}} \right]} - \frac{1}{N_3^{PS} \beta_1 \left[ 1 - \frac{\lambda^2}{N_3^{PS}} - \frac{2\lambda}{\beta_1 \sqrt{N_3^{PS}}} \right]} \right]$$

$$P_{1,8cm}^{PS} = \frac{\mu^{PS} [\lambda - \lambda^{-3}]}{3\lambda_{c,r}} \left[ \beta + \frac{1}{N_8^{PS} \lambda_{c,r}} - \frac{1}{N_8^{PS} \beta \left[ 1 - \lambda_{c,r}^2 - \frac{2\lambda_{c,r}}{\beta} \right]} \right]$$

$$\text{where, } \lambda_{c,r} = \sqrt{\frac{1}{3N_8^{PS}} [\lambda^2 + \lambda^{-2} + 1]}, \beta = \lambda_{c,r} \frac{3 - \lambda_{c,r}^2}{1 - \lambda_{c,r}^2}, \beta_1 = \frac{\lambda}{\sqrt{N_3^{PS}}} \frac{3N_3^{PS} - \lambda^2}{N_3^{PS} - \lambda^2}, \beta_4 = \frac{\lambda^{-1}}{\sqrt{N_3^{PS}}} \frac{3N_3^{PS} - \lambda^{-2}}{N_3^{PS} - \lambda^{-2}}.$$

3 References

1. Amin AFMS (2001) Constitutive modeling of strain-rate dependency of natural and high damping rubbers, PhD Dissertation, Saitama University, Japan
2. Amin AFMS, Wiraguna SI, Bhuiyan AR, Okui Y (2006a) Hyperelasticity model for finite element analysis of natural and high damping rubbers in compression and shear. Journal of Engineering Mechanics ASCE 132(1): 54-64
3. Amin AFMS, Lion A, Sekita S, Okui Y (2006) Nonlinear dependence of viscosity in modeling the rate-dependent response of natural and high damping rubbers in compression and shear: Experimental identification and numerical verification. International Journal of Plasticity 22: 1610-1657
4. Milani G, Milani F (2012) Stretch-stress behavior of elastomeric seismic isolators with different rubber materials: Numerical insight. Journal of Engineering Mechanics ASCE 138(5): 416-428

- 
- 1 5. Treloar LRG (1944) Stress-strain data for vulcanised rubber under various types of deformation. Transactions of the Faraday  
2 Society 40: 59-70
  - 3 6. Treloar LRG, Riding G (1979) A non-Gaussian theory of rubber in biaxial strain. I. Mechanical properties. Proceedings of  
4 the Royal Society of London. A369, 1979, p.261
  - 5 7. Beatty MF (2003) An average-stretch full-network model for rubber elasticity. Journal of Elasticity 70: 65-86
  - 6 8. Miehe C, Göktepe S, Lulei F (2004) A micro-macro approach to rubber-like materials - Part I: The non-affine micro-sphere  
7 model of rubber elasticity. Journal of the Mechanics and Physics of Solids 52: 2617-2660
  - 8 9. Boyce MC, Arruda EM (2000) Constitutive models of rubber elasticity: a review. Rubber Chemistry and Technology 73:  
9 504-523,2000
  - 10 10. Elias-Zuniga A, Beatty MF (2002) Constitutive equations for amended non-Gaussian network models of rubber elasticity.  
11 International Journal of Engineering Science 40: 2265-2294
  - 12 11. Arruda EM, Boyce MC (1993) A three-dimensional constitutive model for the large stretch behavior of rubber elastic  
13 materials. Journal of the Mechanics and Physics of Solids 41: 389-412
  - 14 12. Ogden RW (1972) Large deformation isotropic elasticity - on the correlation of theory and experiment for incompressible  
15 rubberlike solids. Proceedings of the Royal Society of London. Series A, Mathematical and Physical Sciences 326: 565-584
  - 16 13. Ogden RW, Saccomandi G, Sgura I (2004) Fitting hyperelastic models to experimental data. Computational Mechanics 34:  
17 484-502
  - 18 14. Wu PD, van der Giessen E (1992) On improved 3-D non-Gaussian network models for rubber elasticity. Mechanics Research  
19 Communications 19: 427-433
  - 20 15. Kroon M (2011) An 8-chain model for rubber-like materials accounting for non-affine chain deformations and topological  
21 constraints. Journal of Elasticity 102 (2): 99-116, 2011,
  - 22 16. Marckmann G, Verron E (2006) Comparison of hyperelastic models for rubber-like materials. Rubber Chemistry and Tech-  
23 nology 79: 835-858
  - 24 17. Kaliske M, Rothert H (1997) On the finite element implementation of rubber-like materials at finite strains. Engineering  
25 Computations 14: 216-232
  - 26 18. Kaliske M, Heinrich G (1999) An extended tube-model for rubber elasticity: Statistical mechanical theory and finite element  
27 implementation. Rubber Chemistry and Technology 72: 602-632
  - 28 19. Chagnon G, Marckmann G, Verron E (2004) A comparison of the Hart-Smith model with Arruda-Boyce and Gent formu-  
29 lations for rubber elasticity. Rubber Chemistry and Technology 77: 724-735
  - 30 20. Hart-Smith LJ (1966) Elasticity parameters for finite deformations of rubber like materials. Zeitschrift für Angewandte  
31 Mathematik und Physik 17: 608-626
  - 32 21. Flory PJ, Erman B (1980) Theory of elasticity of polymer networks 3. Macromolecules 15(3): 800-806
  - 33 22. Jernigan, RL, Flory PJ (1969) Distribution functions for chain molecules. Journal of Chemical Physics 50: 4185-4201
  - 34 23. Shariff MHB (2000) Strain energy function for filled and unfilled rubber-like material. Rubber Chemistry and Technology  
35 73: 1-18
  - 36 24. Elias-Zuniga A (2006) A non-Gaussian network model for rubber elasticity. Polymer 47: 907-914
  - 37 25. Pucci E, Saccomandi G (2002) A note on the Gent model for rubber-like materials. Rubber Chemistry and Technology  
38 75(5):839-851
  - 39 26. Hossain M, Steinmann P (2013) More hyperelastic models for rubber-like materials: Consistent tangent operator and com-  
40 parative study. Journal of the Mechanical Behaviour of Materials 22(1-2):27-50
  - 41 27. Hossain M, Vu D K, Steinmann P (2012) Experimental study and numerical modelling of VHB 4910 polymer. Computa-  
42 tional Materials Science 59:65-74

- 
- 1 28. Steinmann P, Hossain M, Possart G (2012) Hyperelastic models for rubber-like materials: Consistent tangent operators and  
2 suitability for Treloar's data. *Archive of Applied Mechanics*, DOI:10.1007/s00419-012-0610-z
  - 3 29. Morini B, Porcelli M (2010) TRESNEI, a Matlab trust-region solver for systems of nonlinear equalities and inequalities.  
4 *Computational Optimization and Applications*, DOI: 10.1007/s10589-010-9327-5
  - 5 30. Jerrams S, Murphy N (Editors) Constitutive models for rubber VII. Proceedings of the 7th european conference on consti-  
6 tutive models for rubber, Dublin, Ireland. CRC Press (Taylor & Francis Group), 2012
  - 7 31. Miroshnychenko D, Green WA, Turner DM (2005) Composite and filament models for the mechanical behaviour of elas-  
8 tomeric materials. *Journal of the Mechanics and Physics of Solids* 53:748-770
  - 9 32. Miroshnychenko D, Green WA (2009) Heuristic search for a predictive strain-energy function in nonlinear elasticity. *Inter-  
10 national Journal of Solids and Structures* 46:271-286
  - 11 33. Bechir H, Chevalier L, Idjeri M (2010) A three-dimensional network model for rubber elasticity: The effect of local entan-  
12 glements constraints. *International Journal of Engineering Science* 48: 265-274
  - 13 34. Meissner B, Matejka L (2003) A Langevin-elasticity-theory-based constitutive equation for rubberlike networks and its  
14 comparison with biaxial stress-strain data. Part I. *Polymer* 44: 4599-4510
  - 15 35. Meissner B, Matejka L (2004) A Langevin-elasticity-theory-based constitutive equation for rubberlike networks and its  
16 comparison with biaxial stress-strain data. Part II. *Polymer* 45: 7247-7260
  - 17 36. Itskov M, Ehret AE, Dargazany R (2010) A full-network rubber elasticity model based on analytical integration. *Mathemat-  
18 ics and Mechanics of Solids* 15: 655-671
  - 19 37. Sahu R K, Patra K (2013) Estimation of elastic modulus of dielectric elastomer materials using Mooney-Rivlin and Ogden  
20 Models. *Advanced Materials Research* 685: 331-335