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A comparison of limited-stretch models of rubber elasticity

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ABSTRACT

In this paper we describe various limited-stretch models of non-linear rubber elasticity, each dependent on only the first invariant of the left Cauchy–Green strain tensor and having only two independent material constants. The models are described as limited-stretch, or restricted elastic, because the strain energy and stress response become infinite at a finite value of the first invariant. These models describe well the limited stretch of the polymer chains of which rubber is composed. We discuss Gent's model which is the simplest limited-stretch model and agrees well with experiment. Various statistical models are then described: the one-chain, three-chain, four-chain and Arruda–Boyce eight-chain models, all of which involve the inverse Langevin function. A numerical comparison between the three-chain and eight-chain models is provided. Next, we compare various models which involve approximations to the inverse Langevin function with the exact inverse Langevin function of the eight-chain model. A new approximate model is proposed that is as simple as Cohen's original model but significantly more accurate. We show that effectively the eight-chain model may be regarded as a linear combination of the neo-Hookean and Gent models. Treloar's model is shown to have about half the percentage error of our new model but it is much more complicated. For completeness a modified Treloar model is introduced but this is only slightly more accurate than Treloar's original model. For the deformations of uniaxial tension, biaxial tension, pure shear and simple shear we compare the accuracy of these models, and that of Puso, with the eight-chain model by means of graphs and a table. Our approximations compare extremely well with models frequently used and described in the literature, having the smallest mean percentage error over most of the range of the argument.

1. Introduction

Several strain energy models of rubber elasticity are developed in this paper, some based on positing a form of the strain energy function, some based on statistical mechanical considerations of the polymer chains of which rubber is composed and others based on approximations to the statistical mechanics models. The statistical mechanics models we consider in this paper involve only the first invariant I_1 of the left Cauchy–Green strain tensor. The simplest statistical model is based on Gaussian statistics and leads to the well known neo-Hookean strain energy function which depends linearly on I_1 and on only one material constant, the shear modulus. It is a feature of rubber elasticity that when a specimen of rubber is stretched in any direction a maximum stretch is reached, corresponding to the polymer chains being stretched to their maximum extent. We model this property by requiring the stress response and strain energy to become infinite as this maximum stretch is reached. Such a model of rubber elasticity is said to be a limited-stretch, or restricted elastic model. All the other models we discuss are of this type and, furthermore, depend on only two material constants, a shear modulus and the value I_m of the first invariant when maximum stretch is reached. Gent [13] discusses the relevance of modelling rubber using only the first principal invariant I_1 with its maximum I_m. Horgan and Saccomandi [16] have shown that this approach of limiting chain extensibility may be used to model biological material.

All the models considered in this paper are freely jointed chain (FJC) models. The alternative worm-like chain (WLC) model is discussed briefly at the end of Section 3.

We consider the strain energy and stress response for each of the following models: Gent [11], Beatty [3], Van der Waals [22] and Warner [31]. None of these is directly related to statistical mechanics but each has limited stretch and depends on only two elastic constants.

More sophisticated statistical modelling also leads to limited-stretch models with two elastic constants, each depending on the inverse Langevin function and deriving its limited-stretch

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behaviour from the singularity of this function. Kuhn and Grün [23] used statistical mechanics to derive an expression for the strain energy function of a single polymer chain which involved the inverse Langevin function. A similar approach has been used to develop network models based on cell structures, including the James and Guth [21] three-chain model, the Wang and Guth [30] four-chain model and the Arruda and Boyce [1] eight-chain model.

When using any of the above models an approximation to the inverse Langevin function is required. Perhaps the simplest approximations are those obtained by truncating the Taylor series. However, many terms of the Taylor series are needed to approach convergence, see Itskov et al. [19,20] for further discussion. Most approximations involve the Taylor series, such as the method of Padé approximants or further approximations to these. Horgan and Saccomandi [17] note that such methods may be used to capture correctly the real singularities of the inverse Langevin function. Cohen [8] derived an approximation based on the [3/2] Padé approximant of the inverse Langevin function and Treloar [29] obtained a rational approximation to the inverse Langevin function which is related to the [1/6] Padé approximant of its Taylor series. We present a new model, and a modified Treloar model, which are based on Padé approximants of the reduced Langevin function, a function defined by multiplying out the simple poles of the inverse Langevin function. Puso's [26] model does not appear to depend directly on Padé approximants. We also include a discussion on the additive removal of the real singularities of the inverse Langevin function.

Zúñiga and Beatty [33] and Beatty [2] describe the James and Guth [21] three-chain model, the Arruda and Boyce [1] eight-chain model and the Wu and van der Giessen [32] full network model. Beatty [3] discusses the derivation of the Cohen [8], Treloar [29] and Horgan and Saccomandi [15] approximations and concludes that the approximation of Treloar [29] is the most accurate over the entire range of its argument. Boyce [5] directly compares the Gent and eight-chain models concluding that the eight-chain model gives a better interpretation of the physics of the polymer chain network, though both models provide excellent agreement with experimental data.

This paper is structured as follows. In Section 2 we define the Cauchy stress for an incompressible isotropic elastic material and specialise to the case where the strain energy depends only on the first invariant, I_1, of the left Cauchy–Green strain tensor. We derive expressions for the stress in the following four homogeneous deformations: uniaxial tension, biaxial tension, pure shear and simple shear. In Section 3 we discuss several models for limited-stretch rubber elasticity that are dependent on the first invariant only. These models are: the neo-Hookean, Gent, Beatty, van der Waals and Warner models. The stress response and strain energy are given for each model. In Section 4 we define the Langevin function and its inverse $\mathcal{L}^{-1}(x)$ and present series expansions for them. We introduce a reduced inverse Langevin function $f(x)$ which consists of the inverse Langevin function $\mathcal{L}^{-1}(x)$ with its simple poles removed by multiplying them out. A series expansion for $f(x)$ is also given. In Section 5 we describe several models of limited-stretch rubber elasticity which are based on the inverse Langevin function, namely, the single-chain, three-chain, eight-chain and four-chain models. We note also in this section that Beatty [2] demonstrates an alternative derivation of the eight-chain model without reference to the eight-chain cell structure. We conclude this section with a numerical comparison between the three-chain and eight-chain models. In Section 6 we discuss several models which are based on approximations to the inverse Langevin function. The first model consists of various truncations of the power series of $\mathcal{L}^{-1}(x)$. This series and the series for $f(x)$ play an important role in the further models introduced, namely, those of Cohen, a new model, Treloar's model, a modification of Treloar's model, Puso's model, Indei et al.'s model and a model based on the additive removal of the real singularities of $\mathcal{L}^{-1}(x)$. For most of these models we present the stress response and strain energy. We show that to a high degree of accuracy the eight-chain model may be regarded as a linear combination of the neo-Hookean and Gent models. In Section 7 we give a numerical comparison of the various models, taking as reference the Arruda–Boyce [1] eight-chain model. We first provide a graphical comparison of the neo-Hookean, Gent, Beatty, van der Waals and Warner models with the Arruda–Boyce eight-chain model, comparing the stress responses and strain energies. We then proceed to compare graphically Cohen's model, the new model, Treloar's model and the modified Treloar model with the Arruda–Boyce eight-chain model. We consider the stress response, the strain energy and each of the four homogeneous deformations discussed in Section 2. We also give the mean percentage errors for all these quantities in a table. Finally, there is a discussion of the results in Section 8.

2. Four homogeneous deformations

The Cauchy stress in an incompressible isotropic elastic material is given by

$$\mathbf{T} = -p\mathbf{I} + \beta\mathbf{B} + \beta_{-1}\mathbf{B}^{-1} \tag{1}$$

where p is an arbitrary pressure and $\mathbf{B} = \mathbf{F}\mathbf{F}^{\mathrm{T}}$ is the left Cauchy–Green strain tensor with $\mathbf{F}$ denoting the deformation gradient. The response functions are given in terms of the strain energy W by

$$\beta = 2\frac{\partial W}{\partial I_1}, \quad \beta_{-1} = -2\frac{\partial W}{\partial I_2} \tag{2}$$

where $I_1 = \mathrm{tr}\,\mathbf{B}$ and $I_2 = \mathrm{tr}\,\mathbf{B}^{-1}$ are the first two principal invariants of $\mathbf{B}$. Because of incompressibility the third principal invariant is given by $I_3 = \det \mathbf{B} = 1$. We are assuming no dependence on I_2 and so must take $\beta_{-1} = 0$ and $\beta = \beta(I_1)$. Therefore, throughout this paper, the Cauchy stress (1) reduces to

$$\mathbf{T} = -p\mathbf{I} + \beta\mathbf{B}, \tag{3}$$

where the stress response β is given by Eq. $(2)_1$.

A method alternative to Eq. (3) of designating the stress in an incompressible isotropic elastic material is to take the strain energy to be a symmetric function of the principal stretches, $W = \widehat{W}(\lambda_1, \lambda_2, \lambda_3)$, and observe that the principal Cauchy stresses are given by

$$T_j = -p + \lambda_j \frac{\partial \widehat{W}}{\partial \lambda_j}, \quad \text{for} \quad j = 1, 2, 3, \tag{4}$$

in which the principal stretches are denoted by λ_j, for $j = 1, 2, 3$. In terms of the principal stretches we have

$$I_1 = \mathrm{tr}\,\mathbf{B} = \lambda_1^2 + \lambda_2^2 + \lambda_3^2. \tag{5}$$

We now examine four different homogeneous deformations and in each case compute the stress in terms of the largest principal stretch $\lambda > 1$ and the response function β.

2.1. Uniaxial tension

We consider a uniaxial tension in the 1-direction with corresponding principal stretch $\lambda_1 = \lambda > 1$, so that incompressibility forces the other two principal stretches to be $\lambda_2 = \lambda_3 = \lambda^{-1/2}$. We take $p = \lambda^{-1}\beta$ to ensure the vanishing of the lateral stresses and then the only non-zero stress is the uniaxial tension

$$T_{11}^{\mathrm{uni}}(\lambda) = (\lambda^2 - \lambda^{-1})\beta, \quad I_1 = \lambda^2 + 2\lambda^{-1}. \tag{6}$$