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On Pseudo-Elastic Models for Stress Softening in Elastomeric Balloons

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Abstract: The phenomenon of stress softening observed in the cyclic inflation of spherical balloons or membranes is quantitatively and qualitatively examined. A new measure of the stress softening extent is proposed which correctly captures the main feature of this phenomenon. This measure of the stress softening is related to the relevant response functions in the constitutive models proposed in the literature to describe this effect. Using these relationships, the predictive capability of the theoretical models is examined. It is shown that only those theoretical models which admit a non-monotone character of the stress softening can properly describe this phenomenon.

Keywords: Spherical balloons, membranes, elastomers, cyclic inflation, stress softening.

1 Introduction

It has long been known that the inflation of a toy balloon may provide a good illustration of a peculiar behavior of elastomeric membranes. Typically, in such an inflation test the following sequence of events is recorded: initially the radius of the balloon increases continually until the pressure reaches a maximum at a relatively small deformation. If the balloon is further inflated, the pressure decreases and finally rises again at very large strains. The data illustrating this type of behavior of elastomeric balloons were already presented at the beginning of the last century by Osborne (1909). This peculiar property of balloons results from the combined effects of geometric and material non-linearity and the observed non-monotonous (N-shaped) pressure-radius relationship gives rise to a highly nontrivial stability problem [Müller and Strehlow (2004)].

Over years, the problem of inflation of balloons or spherical membranes has been studied by many authors under the assumption that the deformation is completely reversible, e.g. Hart-Smith (1966) and Alexander (1971). These papers contain

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also interesting experimental data. The review of the earlier studies in this field may be found in Crisp and Hart-Smith (1971), Chen and Healey (1991) and in the more recent study by Gent (1999).

It must be noted, however, that the perfect elasticity is often a very crude assumption. A typical inelastic effect observed in the inflation and subsequent deflation of balloons is that of hysteresis accompanied by stress-softening (the Mullins effect) and residual strains (the permanent set) as it may be seen from data presented by Beatty (1987) and Johnson and Beatty (1995). It is interesting to note that the phenomenon of stress softening of balloons has already been recognized by Sutherland as it may be seen from his letter of 1908 to Osborne (see [Bolton and Rae (1994)]). This phenomenon is also observed in the inflation of certain biological membranes, e.g. van Mastrigt, et al. (1978) and Hughes and Vergara (1978). Therefore, the understanding of the nature and properties of the stress softening phenomenon and its mathematical modeling is of importance not only in the science of elastomers but also in the physiology of natural organs. In fact, the study of inflation of membranes has been largely stimulated by the research in physiology (see e.g. [Frank (1910); Brody and Quigley (1948); Rigato (1967)]).

The phenomenon of stress softening observed in the cyclic inflation-deflation tests of spherical balloons or membranes has been studied by Beatty and Krishnaswamy (2000) within their own theoretical model for the Mullins effect in incompressible elastomers. Very recently, this problem has been considered by Ren (2008) within the concept of pseudo-elasticity, originally proposed by Ogden and Roxburgh (1999) and further developed by Dorfmann and Ogden (2004). However, neither of these two studies presents a comparison of the theoretical results with the available experimental data. Moreover, an analysis of their results shows that the main physical characteristics of the stress softening phenomenon are not properly represented. In particular, the results of Ren (2008) appear to be at marked variance with the experimental data shown in Beatty (1987), the paper to which Ren refers in his study.

In this work a new methodology of analyzing data of cyclic inflation tests of balloons or spherical membranes is presented. It leads to the concept of stress retention giving a measure of the amount of stress softening recorded in every inflation-deflation cycle. This measure provides the quantitative and qualitative representations of the main physical characteristics of the stress softening and serves as basic tool for the study of predictive capabilities of theoretical models of this phenomenon. In order to be able to study various theoretical models proposed in the literature, general theory of the stress softening for the radially symmetric deformation of spherical balloons is first developed. This theory takes into account the residual strains besides the stress softening effect. Next, the developed theory is reduced to the model originally presented by Dorfmann and Ogden (2004). This model contains as a special case the model developed by Ogden and Roxburgh (1999) which is obtained by neglecting the residual strains. The results predicted by these models are compared with the data of Beatty (1987). It is pointed out that the theoretical results show certain divergence from the experimental data. The reasons of this discrepancy are discussed and, finally, the thermodynamic basis of the pseudo-elastic models for the Mullins effect is examined.

2 Cyclic inflation tests - data analysis

Let us consider a thin spherical membrane or balloon of radius R_0 and uniform thickness $h_0 \ll R_0$ in its initial (undeformed) state, and subject to an inflation pressure p. In a typical inflation test, the measurable quantities are the pressure p and volume V of the balloon. If the balloon remains spherical during the inflation process with the deformed radius R and uniform thickness $h \ll R$, then the measurements of V is equivalent to the measurements of the diameter $D = 2R$ of the inflated balloon with volume calculated from the classical formula $V = 4\pi R^3/3$. The initial state of the balloon is characterized by $p = 0$ and the initial volume $V_0 = 4\pi R_0^3/3$.

That the balloon remains spherical during the inflation process implies that the skin of the balloon stretches by the same proportion in all directions. Hence, each surface element undergoes an equibiaxial in-plane extension $\lambda \equiv \lambda_1 = \lambda_2$ equal to the radial stretch given by $\lambda = R/R_0$. The uniform transverse normal stretch λ_3 of the balloon is determined by the change in thickness. Moreover, the change in volume is given by the formula $\upsilon \equiv V/V_0 = \lambda^3$. Thus, the data of a typical inflation test may be represented as $p - \upsilon$ curve, equivalently, as $p - \lambda$ curve.

The data obtained from a typical cyclic inflation experiment is shown in Fig. 1, where the inflation and deflation curves over one cycle are followed by inflation to failure. There is a notable retracing of a similar curve in the deflation phase which includes a relative maximum pressure at a stretch larger than that corresponding to the maximum on the primary inflation curve. The smaller maximum pressure attained in the second inflation is consonant with every day experience in prestretching a balloon prior to its primary inflation.

There are different ways of characterizing the amount of stress softening effect seen in Fig. 1. For example, the change in shape of the pressure-stretch curve before and after prestressing to a fixed elongation may be used as a measure of the stress softening. Another way of characterizing the amount of softening is by means of the energy loss ratio which is defined in terms of energy input on primary stretching and energy recovered on unloading. These two methods were studied in detail by Charrier and Gent (1975), who also derived the general relationship between these

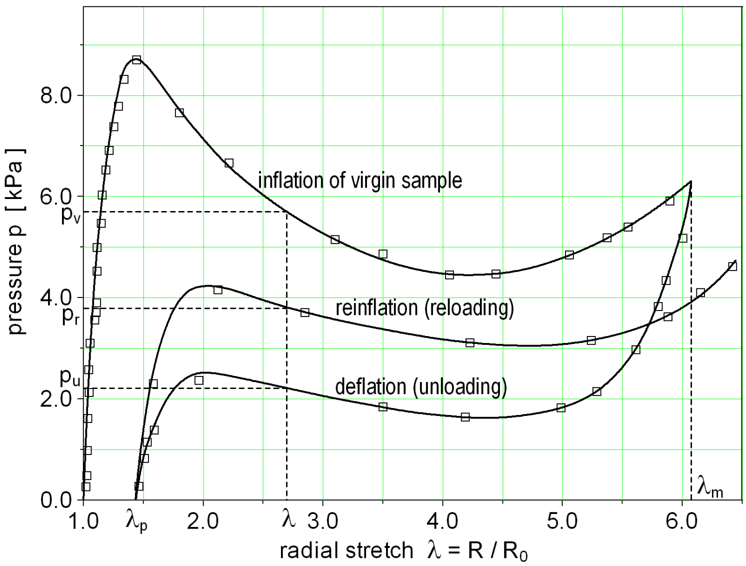

Figure 1: Balloon inflation test - cyclic loading to failure [Beatty (1987)]

two representations of stress softening.

For the purpose of this work, it will be convenient to characterize the amount of stress softening by means of the pressure retention ρ defined as (see Fig. 1)

$$\rho = \rho(\lambda;\lambda_m) = \frac{p_s(\lambda;\lambda_m)}{p_v(\lambda)} \tag{1}$$

for each inflation cycle with the specified value of pre-stretch λ_m. Here p_v is the value of pressure on the primary inflation curve and p_s denotes either the value of pressure p_u on the deflation (unloading) path or the value of the pressure p_r on the reinflation (reloading) path. The data of Fig. 1 replotted as the $\rho - \lambda$ curves are shown in Fig. 2.

As indicated in (1), for each value of pre-stretch λ_m, the stress ratio ρ may be considered as function of the radial stretch λ or the corresponding engineering strain $\varepsilon = \lambda - 1$. Moreover, in order to have the possibility of comparing the amount of softening as measured by ρ for different values of pre-stretch, it is instructive to replot the conventionally represented data in the form giving the stress retention ρ as a function of the dimensionless strain $\xi = (\lambda - 1)/(\lambda_m - 1)$.

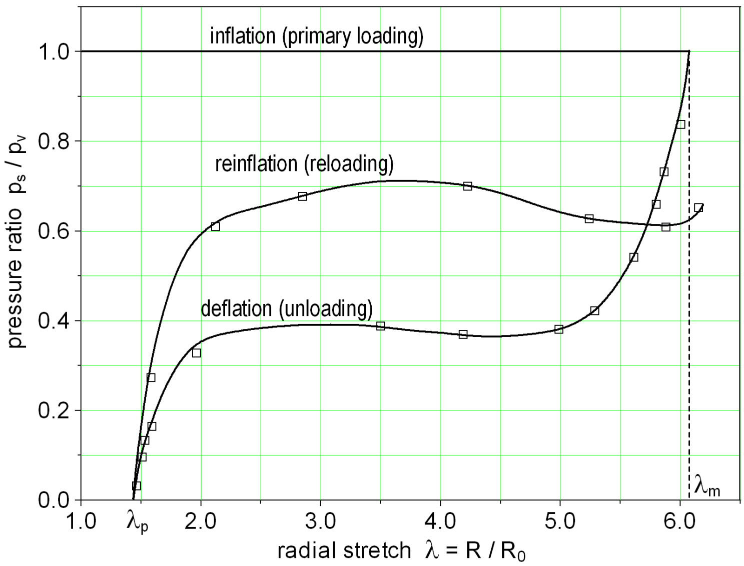

Figure 2: Stress softening in inflation test of balloon represented by stress retention

3 Radially symmetric deformation of balloons

The assumption that the balloon is a spherically symmetric membrane of constant initial thickness and uniform material properties implies that the state of stress is essentially equibiaxial. Thus, the resultant (two-dimensional) true membrane stress tensor N has the form $N = T\mathbf{1}$, where $\mathbf{1}$ is the unit tensor in the tangent plane and T denotes the isotropic extensional force per unit length in the inflated balloon. The equilibrium equation may now be obtained by elementary considerations: equilibrium of a hemispherical portion of the balloon requires that the total edge force be balanced by the total inflation pressure acting over the diametral cross-section of the sphere, that is $2\pi RT = \pi R^2 p$ and hence,

$$\frac{2T}{R} = p\,, \tag{2}$$

which is the famous Laplace law. Let P denote the isotropic, extensional force per unit length in the initial (undeformed) state of the balloon. Then, $2\pi RT = 2\pi R_0 P$ and recalling that $\lambda = R/R_0$, we have $P = \lambda T$ so that the Laplace law (2) may be written as

$$\frac{P}{\lambda^2} = \frac{pR_0}{2}\,. \tag{3}$$

It follows that either the true membrane stress T or the engineering membrane stress P may be used for the analysis.

Because of the assumption $h_0 \ll R_0$, the engineering (first Piola-Kirchhoff) equibiaxial stress $\pi \equiv \pi_1 = \pi_2$ will be nearly uniform across the membrane thickness. Hence, $\pi = P/h_0$ and the Laplace law (3) may be written as

$$\pi = \frac{1}{2}\lambda^2 p^*, \quad p^* \equiv \frac{pR_0}{h_0}, \tag{4}$$

where p^* is the reduced pressure. Thus, the inflation of spherical balloons can be investigated by considering the constitutive relations for equibiaxial stress of elastic or unelastic elastomers. When comparing the theoretical results with the data shown in Fig. 1 it will be assumed $R_0/h_0 = 10^3$.

4 Equibiaxial stress softening models

The constitutive modeling of the stress softening in elastomeric and biological membranes has been considered by Kazakevičiūtė-Makovska (2002) on the basis of a two-dimensional theory of perfectly flexible membranes. For the purpose of this work, however, it is more convenient to use directly the relation (4) and to study the problem of inflation of spherical balloons within the three-dimensional theories reduced to the equibiaxial problems for incompressible elastomer materials.

The constitutive equation for the equibiaxial engineering stress π in the material undergoing stress softening coupled to the permanent set may be assumed in the following general form

$$\pi = \hat{\pi}(\lambda, \alpha, \kappa). \tag{5}$$

Here the response function $\hat{\pi}(\lambda, \alpha, \kappa)$ depends on the equibiaxial stretch λ and two internal variables: the softening variable α and the residual strain variable κ. These two variables must be specified by appropriate evolution laws in consistency with the relevant softening and residual strain criteria. In modeling the behavior of elastomers, it is generally assumed that the response of these materials to cyclic loading depends only on the maximum stretch λ_m previous experienced by the material during the deformation history (see e.g. [Dorfmann and Ogden (2004)] and references cited therein). Accordingly, the evolution laws for the softening and residual strain variables may be assumed in the following general form

$$\alpha = \hat{\alpha}(\lambda; \lambda_m), \quad \kappa = \hat{\kappa}(\lambda; \lambda_m). \tag{6}$$

Introducing the constitutive equation (5) into the Laplace law (4), the reduced pressure p^* is obtained as

$$p^* = \hat{p}^*(\lambda, \alpha, \kappa) = 2\lambda^{-2}\hat{\pi}(\lambda, \alpha, \kappa) \tag{7}$$

with the evolution laws for α and κ given by (6).

Within the constitutive laws (5)-(7), two broad classes of theoretical models for the stress softening may now be distinguished. The first class of models, which is the subject of this paper, is based on the assumption that both variables α and κ take zero value during the primary inflation of a virgin balloon, while they evolve according to specified evolution laws (6) upon deflation and subsequent reinflation provided that the deformation is smaller than the previous one. Since the maximum stretch on the primary inflation path is equal the current stretch, $\lambda_m = \lambda$, this assumption requires that

$$\hat{\alpha}(\lambda;\lambda) = 0, \quad \hat{\kappa}(\lambda;\lambda) = 0 \tag{8}$$

for all $\lambda \in [1, \lambda_m]$ and every pre-stretch λ_m. By implication, the constitutive equation (5) reduces to the classical law of non-linear elasticity

$$\pi = \hat{\pi}_E(\lambda) = \hat{\pi}(\lambda, 0, 0) = \frac{1}{2}\hat{w}'(\lambda)\,. \tag{9}$$

Here $w = \hat{w}(\lambda)$ is the strain energy function specified for the equibiaxial deformation and a prime indicates the derivative with respect to λ. Correspondingly, the primary inflation of the virgin balloon is modeled by the constitutive law of non-linear elasticity

$$p^* = \hat{p}^*_E(\lambda) = 2\lambda^{-2}\hat{\pi}_E(\lambda)\,, \tag{10}$$

where the response function $\hat{\pi}_E(\lambda)$ is given by (9).

It follows that within this class of models the behavior of a virgin balloon during the primary inflation is completely described by the elastic constitutive law (9) and the deflation (unloading) and subsequent reinflation (reloading) of the balloon are defined by the general constitutive law (5). To this class belongs the model developed by Dorfmann and Ogden (2004) as well as its special version without residual strains taken into account first proposed Ogden and Roxburgh (1999). These two special models are considered below.

The second class of models, which may be developed within the general constitutive equations (5)-(7), is based on the assumption that the variables α and κ evolve along the primary loading path (the virgin pressure-deformation curve) according to a specified rule while they take constant values upon unloading (deflation) and subsequent reloading (reinflation) provided that the actual stretch does not exceed its maximum value achieved in the previous deformation. The model proposed by Beatty and Krishnaswamy (2000) with the residual strains neglected belongs to this class, but this model will not be studied in this paper.

4.1 Idealized stress softening models (without residual strains)

The theoretical model for the idealized Mullins effect, i.e. without residual strains taken into account, has been developed by Ogden and Roxburgh (1999) within the new concept of pseudo-elasticity. When specified for the equibiaxial deformation, their model yields the stress-strain relation in the form

$$\pi = \hat{\pi}(\lambda, \alpha, \kappa) = (1 - \hat{\alpha}(\lambda; \lambda_m))\hat{\pi}_E(\lambda) \tag{11}$$

with the evolution law for the softening variable α assumed by Dorfmann and Ogden (2004) in the form

$$\alpha = \hat{\alpha}(\lambda; \lambda_m) = \tilde{\alpha}(w; w_m) = \frac{1}{r} \tanh\left(\frac{w_m - w}{m\mu}\right). \tag{12}$$

In (11), $\hat{\pi}_E(\lambda)$ is the response function for the stress on the primary inflation path and it is given by the classical elastic law (9). In (12), $r > 0$ and $m > 0$ are dimensionless positive material parameters, and $\mu > 0$ is the shear modulus. Moreover, $w = \hat{w}(\lambda)$ denotes the value of elastic energy at the current stretch and $w_m = \hat{w}(\lambda_m)$ its maximum value attained at the prestretch λ_m.

The Ogden-Roxburgh (1999) model may be considered as a special case of the general theory presented in Section 4. In particular, it is seen that the evolution law (12) satisfies the condition $(8)_1$. By implication, the softening variable in the Ogden-Roxburgh (1999) theory is directly related to the experimentally based variable defined in (1), namely, we have

$$\rho(\lambda; \lambda_m) = 1 - \hat{\alpha}(\lambda; \lambda_m). \tag{13}$$

Accordingly, the softening function $\hat{\alpha}(\lambda; \lambda_m)$ in the constitutive equation (11) may be determined directly from experimental data.

4.2 Stress softening model including residual strains

Dorfmann and Ogden (2004) extended the original theory due to Ogden-Roxburgh (1999) by taking into account the residual strains besides the stress softening. Their theory involves two internal variables, as in the theory presented in Section 4, and for equibiaxial deformation yields the following constitutive equation for the equibiaxial engineering stress

$$\begin{aligned} \pi &= \hat{\pi}(\lambda, \alpha, \kappa) \\ &= (1 - \hat{\alpha}(\lambda; \lambda_m))\hat{\pi}_E(\lambda) + \hat{\kappa}(\lambda; \lambda_m)\hat{\pi}_P(\lambda). \end{aligned} \tag{14}$$

Here the response function $\hat{\pi}_E(\lambda)$ for the stress on the primary loading path is given by the elastic law (9) and the response function $\hat{\pi}_P(\lambda)$ is defined by

$$\hat{\pi}_P(\lambda) = \frac{1}{2}\hat{n}'(\lambda)\,, \tag{15}$$

where $\hat{n}(\lambda)$ denotes the equibiaxial energy function which characterizes the residual strains. In Dorfmann and Ogden (2004), the evolution law for the residual strain variable κ was assumed in the form

$$\kappa = \hat{\kappa}(\lambda;\lambda_m) = \tilde{\kappa}(w;w_m) = 1 - C_0^{-1}\tanh\left\{\left(\frac{w}{w_m}\right)^{\beta(w_m)}\right\}. \tag{16}$$

Here $C_0 = \tanh(1)$, $w = \hat{w}(\lambda)$ and

$$\beta(w_m) = b_0 + b_1\mu^{-1}w_m\,. \tag{17}$$

The residual strain energy for the general three-dimensional state of deformation is taken in the form [Dorfmann and Ogden (2004)]

$$N = \hat{N}(\lambda_1,\lambda_2,\lambda_3) = \frac{1}{2}\left\{\nu_1(\lambda_1^2-1)+\nu_2(\lambda_2^2-1)+\nu_3(\lambda_3^2-1)\right\}, \tag{18}$$

with the material parameters ν_i, $i = 1,2,3$, assumed to depend on the maximum stretches λ_i^m in the respective directions, i.e.

$$\nu_i = \hat{\nu}_i(\lambda_i^m), \quad i = 1,2,3\,. \tag{19}$$

The residual strain energy function (18) together with specific laws (19) determine the residual strains.

In Dorfmann and Ogden (2004), the uniaxial state of deformation has only been analyzed in detail. In this case, $\lambda \equiv \lambda_1$ and $\lambda_2 = \lambda_3 = \lambda^{-1/2}$ due to incompressibility assumption, so that the residual strain energy (18) simplifies to the form

$$\hat{n}(\lambda) \equiv \hat{N}(\lambda,\lambda^{-1/2},\lambda^{-1/2}) = \frac{1}{2}\left\{\nu_1(\lambda^2-1)+(\nu_2+\nu_3)(\lambda^{-1}-1)\right\}. \tag{20}$$

Correspondingly, the response function for the residual stress is obtained in the form

$$\hat{\pi}_P(\lambda) = \hat{n}'(\lambda) = \nu_1\lambda - \frac{1}{2}(\nu_2+\nu_3)\lambda^{-2} \tag{21}$$

with the material parameters assumed by Dorfmann and Ogden (2004) in the form

$$\nu_1 = \hat{\nu}_1(\lambda_m) = c_0\mu \{1 - c_1 \tanh(c_2(\lambda_m - 1))\} \tag{22}$$

and

$$\nu_2 = \nu_3 = \frac{1}{2} c_0 \mu \,. \tag{23}$$

For the equibiaxial deformation, $\lambda \equiv \lambda_1 = \lambda_2$ and $\lambda_3 = \lambda^{-2}$, and the corresponding residual strain energy is obtained as

$$\hat{n}(\lambda) \equiv \hat{N}(\lambda, \lambda, \lambda^{-1}) = \frac{1}{2} \left\{ (\nu_1 + \nu_2)(\lambda^2 - 1) + \nu_3(\lambda^{-4} - 1) \right\} . \tag{24}$$

Moreover, for this state of deformation, it is more reasonable to assume that $\nu \equiv \nu_1 = \nu_2$ so that the material parameters are obtained in the form

$$\nu = \nu(\lambda_m) = c_0 \mu d(\lambda_m), \quad \nu_3 = \frac{1}{2} c_0 \mu \,, \tag{25}$$

where the new parameter $d(\lambda_m)$ is defined by

$$d(\lambda_m) = 1 - c_1 \tanh(c_2(\lambda_m - 1)) \,. \tag{26}$$

By implication, the constitutive law for the equibiaxial residual stress is obtained in the form

$$\pi_P = \hat{\pi}_P(\lambda; \lambda_m) = \frac{1}{2} \hat{n}'(\lambda) = c_0 \mu \left\{ d(\lambda_m)\lambda - \frac{1}{2} \lambda^{-5} \right\} . \tag{27}$$

Moreover, since $\hat{\pi}_E(\lambda) > 0$ for all $\lambda > 1$, the constitutive equation (14) can be written as

$$\pi = \hat{\pi}(\lambda; \lambda_m) = (1 - \hat{\alpha}(\lambda; \lambda_m) + \hat{\kappa}(\lambda; \lambda_m)\zeta(\lambda; \lambda_m))\, \hat{\pi}_E(\lambda) \,, \tag{28}$$

where the new response function $\zeta(\lambda; \lambda_m)$ is defined by

$$\zeta(\lambda; \lambda_m) = \frac{\hat{\pi}_P(\lambda; \lambda_m)}{\hat{\pi}_E(\lambda)} \,. \tag{29}$$

The comparison of the constitutive law (28) with the definition (1) shows that

$$\rho = \hat{\rho}(\lambda; \lambda_m) = 1 - \hat{\alpha}(\lambda; \lambda_m) + \hat{\kappa}(\lambda; \lambda_m)\zeta(\lambda; \lambda_m) \,. \tag{30}$$

This relation makes it possible to identify the relevant response functions in Dorfmann-Ogden model directly from the data analysis presented in Section 2 (see Fig. 4).

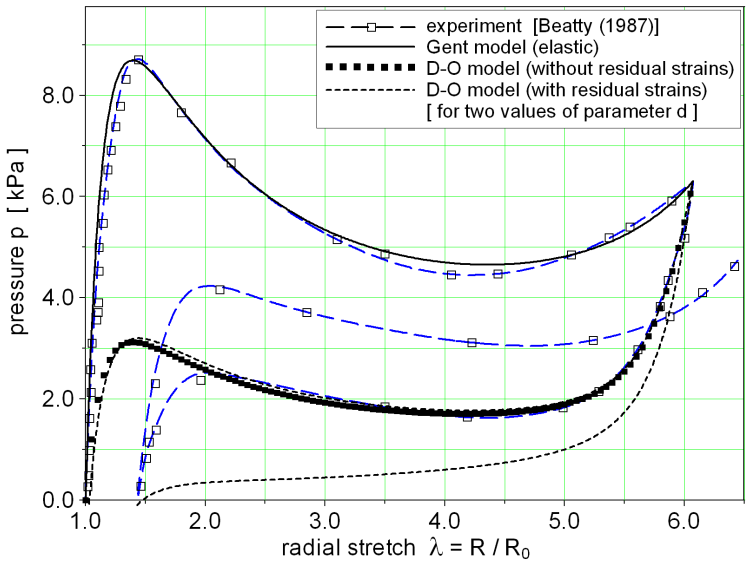

Figure 3: The comparison of the experimental data [Beatty (1987)] with Dorfmann Ogden model predictions

5 Comparison of model predictions with data

Within the pseudo-elastic models for the Mullins effect (without and with residual strains taken into account) studied in this paper, the elastic strain energy function may be used in any form proposed in the literature. Moreover, both models have this convenient property that the material constants appearing in the elastic strain energy function are completely determined from the data for the primary inflation curve. As in Ren (2008), the theoretical results presented below were obtained for the Gent model (see Appendix) with just two elastic constants, the shear modulus $\mu > 0$ having the physical dimension of the Young modulus and the dimensionless parameter J_m. In modeling the data shown in Fig. 1, the value of μ was determined by fitting the maximum pressure $p_{\max}$ and the value of J_m was evaluated from the condition of fitting the value of pressure at the maximum stretch λ_m. This procedure gives $\mu = 3.47\ MPa$ and $J_m = 111$. It is interesting to note that the parameter J_m has only minor influence on the predicted value of the maximum pressure.

Once the elastic constants are determined, there remain to evaluate the values of the material parameters appearing in the evolution laws (12) and (16), and in the residual strain energy (18). It follows from the definition (1) and the derived relation (30) that these parameters are entirely determined by the shape of deflation (or

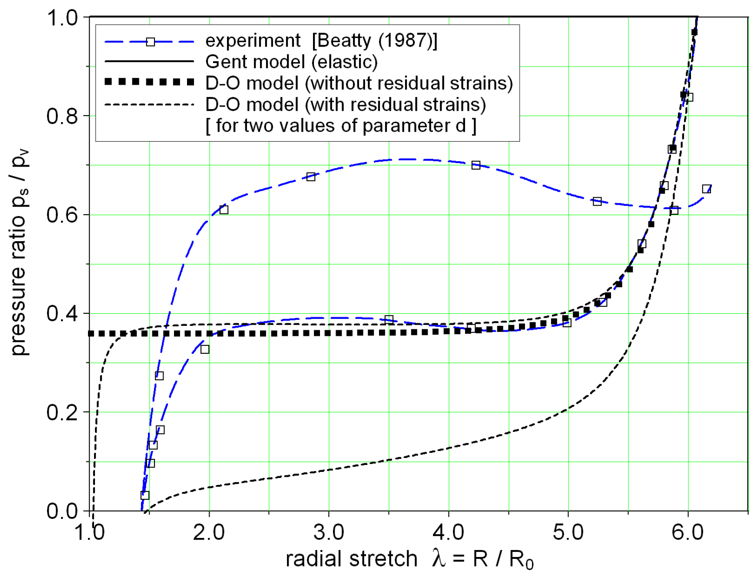

Figure 4: The comparison of the experimental data and theoretical results plotted as pressure ratio vs stretch curve

reinflation) curve.

Considering first the model without residual strains, the material parameters r and m in the evolution laws (12) were determined by fitting the slope of the deflation curve at the maximum stretch λ_m and the maximum value of pressure on this curve. The material parameters determined by this procedure are $r = 1.56$ and $m = 28.4$.

For the model with residual strains, five additional material constants must be determined, b_0 and b_1 appearing in the evolution law (16) through the parameter $\beta(w_m)$ defined in (17), c_0 appearing in the expressions (25) and two constants c_1 and c_2 in the definition (26) of the parameter $d(\lambda_m)$. Recalling that $w_m = w(\lambda_m)$, it becomes clear that for every pre-stretch λ_m only parameters $\beta(w_m)$ and $d(\lambda_m)$ can be determined. In order to determine the values of b_0, b_1 and c_1, c_2, the data must given for at least few inflation cycles with different values of the pre-stretch λ_m. In the analysis of the single inflation cycle only the constant c_0 and two resulting parameters $\beta(w_m)$ and $d(\lambda_m)$ need to be determined.

A comparison of the data [Beatty (1987)] with the results of the Dorfmann-Ogden models with and without residual strains is shown in Fig. 3. These results were obtained for $c_0 = 0.4$ and $\beta(w_m) = 18.2$ as in Ren [2008], and two values of the parameter $d(\lambda_m)$ illustrating the predictive capabilities of the Dorfmann-Ogden model: $d(\lambda_m) = 0.1$ giving the results close to that obtained with the model without residual strains and $d(\lambda_m) = -1.6$ for which the solution fits the experimental value of the residual strain.

It is seen in Fig. 3 that the primary inflation curve of the virgin balloon is very accurate modeled by the Gent elastic model. The stress softening behavior of the balloon is also reasonably well represented by the Dorfmann-Ogden theory with and without residual strains, a far better than this could be seen in the Ren paper (compare the results presented in Fig. 3 with the results shown in Fig. 3 and Fig. 5 in Ren (2008)). It may be noted that Ren [2008] has used the constitutive relations (14), (16) and (17) together with the relations (21), (22) and (23) derived in Dorfmann-Ogden (2004) for the uniaxial and not for the equibiaxial state of deformation. It turns out, however, that this has minor influence on the solutions.

The discrepancies between the theoretical results and experimental data seen in Fig. 3 are the consequence of the monotonic property of the constitutive functions assumed in the evolution laws (12) and (16). This follows from the basic relations (13) and (30) between the measure of stress softening defined in (1) and the evolution laws in the Dorfmann-Ogden theory. It is clearly seen in Fig. 4, where the data and results of Fig. 3 are presented as pressure ratio vs stretch relations, that these constitutive functions must be non-monotonic. In both figures, the results obtained using the Dorfmann-Ogden model with permanent set are shown for two sets of material parameters. For the one set of parameters, the residual strain is very accurately modeled while for the other set the stress softening is more correctly modeled.

6 Energy considerations and discussion

Although the theoretical modeling of the Mullins effect may be based directly on the stress-deformation relation of the general form (5) without any reference to energy considerations (e.g. [Beatty and Krishnaswamy (2000)]), the modern theories of continuum mechanics require that the relevant constitutive equations are compatible with the laws of thermodynamics. In the purely mechanical theories, these laws reduce to the dissipation inequality (see [Kazakevičiūtė-Makovska and Kačianauskas (2004)] and references cited therein) which in the case of equibiaxial deformation takes the form

$$\delta \equiv 2\pi\dot{\lambda} - \dot{\phi} \geq 0. \tag{31}$$

Here $2\pi\dot{\lambda}$ is the stress power density and ϕ is denotes the equibiaxial energy potential identified as a free energy function for isothermal processes. Moreover, the superimposed dot stands for the time derivative. In consistency with the constitutive law for the equibiaxial stress (5), the energy potential is assumed in the form

$\phi = \hat{\phi}(\lambda, \alpha, \kappa)$. Taking the time derivative of ϕ and substituting the resulting expression together with the stress-deformation relation (5) into (31), one gets

$$\delta \equiv \left(2\hat{\pi}(\lambda, \alpha, \kappa) - \partial_\lambda \hat{\phi}(\lambda, \alpha, \kappa)\right)\dot{\lambda} - \left(\partial_\alpha \hat{\phi}(\lambda, \alpha, \kappa)\dot{\alpha} + \partial_\kappa \hat{\phi}(\lambda, \alpha, \kappa)\dot{\kappa}\right) \geq 0\,. \tag{32}$$

It follows that $\phi = \hat{\phi}(\lambda, \alpha, \kappa)$ serves as the potential for the response function for the equibiaxial stress,

$$\hat{\pi}(\lambda, \alpha, \kappa) = \frac{1}{2}\partial_\lambda \hat{\phi}(\lambda, \alpha, \kappa)\,, \tag{33}$$

and the dissipation inequality (32) reduces to the form

$$\delta = -\left(\partial_\alpha \hat{\phi}(\lambda, \alpha, \kappa)\dot{\alpha} + \partial_\kappa \hat{\phi}(\lambda, \alpha, \kappa)\dot{\kappa}\right) \geq 0\,. \tag{34}$$

This reduced dissipation inequality imposes the restrictions on the evolution laws for the variables α and κ, and thus on the response functions $\hat{\alpha}(\lambda;\lambda_m)$ and $\hat{\kappa}(\lambda;\lambda_m)$ in the evolution laws (6). Further possible limitations on the nature of the response functions may only be derived for particular classes of models.

In order to illustrate this point, let us consider the three-dimensional damage-type model of the idealized Mullins effect (without residual strains) developed by Chagnon et al. (2004). When reduced to the equibiaxial deformation, this type of model is based on the the constitutive equation for the energy $\phi = \hat{\phi}(\lambda, \alpha)$ in the special form

$$\phi = \hat{\phi}(\lambda, \alpha) = (1 - \alpha)\hat{w}(\lambda)\,, \tag{35}$$

where $w = \hat{w}(\lambda)$ is the strain-energy of a perfectly elastic material. The assumption (35) together with the general evolution law $(6)_1$ for the softening variable α gives the constitutive equation for the biaxial stress in the form (11). Moreover, $\partial_\alpha \hat{\phi}(\lambda, \alpha) = -\hat{w}(\lambda)$ and since $\hat{w}(\lambda) \geq 0$ by the classical assumption of the non-linear elasticity, the reduced dissipation inequality (34) requires that $\dot{\alpha} \geq 0$. Taking further into account that for this class of models the evolution of the softening variable is given by $(6)_1$, we have

$$\dot{\alpha} = (\partial_\lambda \hat{\alpha}(\lambda;\lambda_m))\,\dot{\lambda} \geq 0\,. \tag{36}$$

For $\dot{\lambda} > 0$ with $1 \leq \lambda \leq \lambda_m$, the inequality (36) implies that $\partial_\lambda \hat{\alpha}(\lambda;\lambda_m) \geq 0$. Thus the softening function $\hat{\alpha}(\lambda;\lambda_m)$ must be a monotonic non-decreasing function of λ for every value of the pre-stretch λ_m.

It is a different matter, however, with the pseudo-elastic models studied in this work. The theory originally formulated by Ogden and Roxburgh (1999) and the extended theory due to Dorfmann and Ogden (2004) which accounts for residual strains are based on energy considerations with the ad hoc assumption that the pseudo-energy potential $\phi = \hat{\phi}(\lambda, \alpha, \kappa)$ satisfies the additional conditions

$$\partial_\alpha \hat{\phi}(\lambda, \alpha, \kappa) = 0, \quad \partial_\kappa \hat{\phi}(\lambda, \alpha, \kappa) = 0\,. \tag{37}$$

These assumptions imply that the reduced dissipation inequality (34) is satisfied identically independently of a particular form of $\phi = \hat{\phi}(\lambda, \alpha, \kappa)$. As a result, there are no a priori restrictions on the admissible forms of the evolutions laws (6) which may be assumed in this class of theoretical models. In particular, the monotonic non-decreasing character of $\hat{\alpha}(\lambda; \lambda_m)$ with respect to the stretch λ for any value of pre-deformation λ_m is not implied by the dissipation inequality, although this is assumed in Ogden and Roxburgh (1999) and Dorfmann and Ogden (2004) models.

In conclusion, the results of this paper may be summarized by stating that the predictive capability of the studied models for the Mullins effect may be improved by admitting the non-monotone forms of the evolution laws. Moreover, the proposed measure ρ of the stress softening extent provides a convenient way of comparing data and theoretical results for the cyclic inflation of spherical balloons. This has been shown in details for the deflation (unloading) curve. The same methodology may be applied to the curve in the reinflation (reloading) phase. It may be noted that using the relations (4) between the pressure and the equibiaxial engineering stress, the measure of stress softening (1) is obtained as $\rho = \pi_s/\pi_v$. Thus, this method of characterizing the amount of stress softening is essentially equivalent to the measure studied by Kazakevičiūtė-Makovska (2007) in the case of uniaxial cyclic deformation.

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Appendix

The elastic strain energy function proposed by Gent (1999) has the form

$$W = W(I_1) = -\frac{\mu}{2} J_m \ln\left(1 - \frac{(I_1 - 3)}{J_m}\right),$$

where μ and J_m are the material constants, and I_1 is the first strain invariant. In the case of equibiaxial deformation of incompressible materials, $\lambda \equiv \lambda_1 = \lambda_2$ and

$\lambda_3 = \lambda^{-2}$ so that

$$I_1(\lambda) = 2\lambda^2 + \lambda^{-4}$$

and the equibiaxial energy function is obtained as

$$\hat{w}(\lambda) \equiv W(I_1(\lambda)) = -\frac{\mu}{2} J_m \ln\left(1 - \frac{(I_1(\lambda) - 3)}{J_m}\right).$$

Both elastic constants in the Gent model have unique physical meaning, $\mu > 0$ is the shear modulus and the dimensionless parameter J_m represents the limiting value for the strain invariant $I_1 - 3$ corresponding to the deformation when the polymer network is fully stretched.