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Modelling the Mullins effect using damage mechanics: efficiency and limitations

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ABSTRACT: The present paper reports on the use of Continuum Damage Mechanics to describe the Mullins effect in elastomers. Thermodynamics of rubber-like hyperelastic materials with isotropic damage is considered. Since it is demonstrated that stress-softening is not a strictly speaking damage phenomenon, the limitations of the approach are highlighted. Moreover, connections with two-network-based constitutive models proposed by other authors are exhibited through the choice of both the damage criterion and the measure of deformation. It is shown that the present model agrees qualitatively well with experiments except for the strain-hardening phenomenon. Finally, the numerical implementation highlights the influence of loading paths on material response, that proves the importance of considering the Mullins effect in industrial design.

1 INTRODUCTION

It is well known that rubber-like materials exhibit a strongly non-linear behaviour characterised by large strain and a non-linear stress-strain response under static conditions. Moreover, this behaviour is timedependent as demonstrated by relaxation or creep experiments. Finally, under cyclic loading conditions, both hysteresis and stress-softening phenomena are observed. The hysteretic behaviour may be related to viscoelasticity or plasticity, and is characterised by different loading and unloading stress-strain paths during a cycle. The stresssoftening phenomenon, also called Mullins effect, is characterised by a consequent loss of stiffness during first cycles of fatigue experiments (see Harwood et al. 1967 and the references therein). As shown by Besdo & Ihlemann (2001), and Kaliske & Domscheit (2001), this phenomenon highly affects the mechanical response of rubber parts

The aim of the present paper is to demonstrate that Continuum Damage Mechanics (denoted CDM through the rest of the paper) can be an efficient tool to model the Mullins effect in rubber, if it is carefully employed. Our work is restricted to hyperelasticity with isotropic damage. In Section 2, a brief review of phenomenological models for the Mullins effect is proposed and the relevance of the CDM theory as applied to the Mullins effect is discussed. The thermodynamical framework of hyperelasticity with damage is derived in Section 3. The similarity of CDM and two-network or twophase approaches is demonstrated. The emphasise is laid on the choice of the damage criterion. Section 4 is devoted to experimental and numerical results. Finally, concluding remarks are given in Section 5.

2 PHENOMENOLOGICAL MODELS FOR THE MULLINS EFFECT

2.1 Two-network or two-phase approaches

The first approach used to develop constitutive equations for the Mullins effect is based on the twonetwork theory of Green & Tobolsky (1946) and the two-phase model of Mullins & Tobin (1957).

Both concepts postulate that the polymer network evolves with deformation: parts of the network are broken and others are reformed under loading. So, Rajagopal & Wineman (1992) proposed a general theory for materials under strain induced changes. microstructural Their approach is successfully applied to the Mullins effect by Huntley et al. (1996, 1997). Authors considered that the stress should be corrected by a scalar reformation function that depends on a given measure of deformation (expressed as a scalar function of principal stretch invariants). Similarly, Beatty & Krishnaswamy (2000) derived a constitutive equation that generalised the previous works of Johnson & Beatty (1993a, b, 1995). In this work, the two-phase theory is considered, and the transformation of hard regions into soft regions is

controlled by an accommodation function that depends on the maximum strain state previously endured by the material. A different form of this function was recently proposed by Elias-Zuniga & Beatty (2002).

2.2 Continuum Damage Mechanics approaches

Many authors used the thermodynamical framework of Continuum Damage Mechanics to model the Mullins effect. CDM was first introduced by Kachanov (1958), and the general theory was derived later by Lemaitre & Chaboche (1985) for metallic materials. This theory was successfully applied to different classes of materials such as metals, composites and concrete.

Recently, CDM was extended to the case of elastomers in order to simulate the Mullins effect. The first paper on the subject is due to Gurtin & Francis (1981) who proposed an one-dimensional hyperelastic damage model for solid propellants. Later, the general three-dimensional case was studied by Simo (1987) who derived a large strain viscoelastic constitutive equation with damage for rubber-like materials. This model was improved by the use of microscopic considerations (Govindjee & Simo 1991, 1992). More recently, several authors developed phenomenological hyperelastic models with damage to describe the Mullins effect in engineering applications (De Souza Neto et al. 1994, Miehe 1995, Kaliske & Rothert (1999), Miehe & Keck 2000, Bikard & Desoyer 2001, Aubard et al. 2002). Finally, let us mention the recent work of Ogden & Roxburgh (1999), used by Muhr et al. (1999) and extended to anisotropy by Holzapfel et al. (1999) that differs from the aforementioned theories because the accommodation function (that depends on the maximum strain energy endured by the material) is activated only on unloading paths.

To conclude this brief review, it should be noticed that constitutive equations based on network evolution are more specific to polymers than CDM models, but their thermodynamical framework is not as well-defined.

2.3 Is the Mullins effect a damage phenomenon?

The application of CDM to the Mullins effect necessitates some remarks. Originally, damage mechanics was based on the definition of effective stress. This theory considers that, under loading, the material surface on which internal forces apply is decreasing because of the emergence of microdefects and micro-voids. In regards with these physical considerations, three consequences are induced for the damage evolution:

- 1. the damage cannot decrease during material life because micro-defects and micro-voids can not disappear,
- 2. the damage evolution differs depending on loading conditions: under tension both micro-defects and micro-voids take place, and under compression only micro-defects are activated because micro-voids are closed,
- 3. the damage evolves until occurrence of a sufficiently large crack, because the active surface tends to a non-zero threshold value determined experimentally.

Nevertheless, in the case of the Mullins effect in elastomers, these three assertions are proved false. More precisely, the Mullins effect is not a damage but a stress-softening phenomenon. It is due to the rearrangement of the polymer network under deformation when some links between chains, or between chains and reinforcement particles are broken (Bueche 1961, 1962). So the three previous assertions can be refuted:

- 1. it was proved experimentally that the Mullins effect recovers with time and that this recovery is highly accelerated by annealing (Mullins 1969). So, the damage may decrease,
- 2. as the Mullins effect is a consequence of the rearrangement of the network, the damage evolution might not differ in regards with deformation modes,
- 3. it is mainly recognised that the rupture in rubber parts is not directly related to stress-softening. Therefore, high values of the damage parameter should not be considered as a criterion for rupture or occurrence of cracks.

Considering these three remarks, it should be concluded that CDM can be used to model the Mullins effect in elastomers with some restrictions and carefulness.

3 CDM AS APPLIED TO RUBBER-LIKE MATERIALS

3.1 Constitutive model

Consider an isotropic, homogeneous and incompressible rubber-like material. This material is considered hyperelastic and subjected to isotropic damage. It is defined by the existence of a strain energy function, which depends on the deformation gradient \mathbf{F} , and on a scalar damage variable *d*. This variable characterises the elastic stress-softening of the material. Taking into account the objectivity requirement, the isotropy and the incompressibility, the strain energy function can be written as:

$$W = W(I_1, I_2, d) \tag{1}$$

where I_1 and I_2 are the two first principal invariants of the left Cauchy-Green strain tensor **B**=**F F**^T, defined by:

$$I_1 = \text{tr}\mathbf{B} \text{ and } I_2 = \frac{1}{2} [I_1^2 - \text{tr}(\mathbf{B}^2)]$$
 (2)

Considering now that the effective Cauchy stress σ_0 , namely the stress that acts on the damaged material, is related to the applied Cauchy stress by (see for example Lemaitre & Chaboche (1985)):

$$\boldsymbol{\sigma}_0 = \frac{\boldsymbol{\sigma}}{1-d} \tag{3}$$

then, the strain energy function can be considered as the product of the surface reducing parameter 1-dand the strain energy function of the virgin undamaged material W_0 :

$$W(I_1, I_2, d) = (1 - d)W_0$$
(4)

In order to establish the laws of state, the Clausius-Duhem inequality is considered and yields (Miehe 1995):

$$(\mathbf{\sigma} + p\mathbf{I}): \mathbf{D} - \dot{W} \ge 0 \tag{5}$$

in which **D** is the rate of deformation tensor. In this equation, $p\mathbf{I}$ is an arbitrary spherical tensor due to incompressibility. After some algebraic manipulations, Equation 5 gives:

$$\boldsymbol{\sigma} = -p\mathbf{I} + 2\left(1 - d\right)\mathbf{B}\frac{\partial W_0}{\partial \mathbf{B}} \quad \text{and} \quad W_0 \dot{d} \ge 0 \tag{6}$$

The left-hand side equation is the constitutive equation that relates the current damaged stress to the damage parameter, the strain energy function of the virgin material and the current strain state. The right-hand side inequality shows that the damage process is dissipative and that the thermodynamic force associated with the damage variable is $-W_0$.

Next, in order to completely define the damaged hyperelastic constitutive equation, the evolution equation of the damage variable has to be established. It is mainly recognised that the stress-softening exclusively depends on the maximum deformation state endured previously by the material. Therefore, a measure of the deformation state and its maximum should be defined. This measure depends on both the deformation state and the time, and is denoted α . Its maximum is:

$$\alpha_m(t) = \max_{\tau \le t} \alpha(\tau) \tag{7}$$

Thus, the damage criterion reduces to a sub-region of the deformation space given by (Simo & Hughes 1998):

$$\phi = \alpha(t) - \alpha_m(t) \le 0 \tag{8}$$

Considering that the damage parameter should increase if, and only if the current deformation state is a maximum state and is in a loading direction from a damage state, an irreversible evolution equation for the damage can be proposed as follow:

$$\dot{d} = \begin{cases} h(\alpha)\dot{\alpha} & \text{if } \phi = 0 \text{ and } \frac{\partial \phi}{\partial \mathbf{B}} : \dot{\mathbf{B}} > 0\\ 0 & \text{otherwise} \end{cases}$$
(9)

where h characterises the evolution of the damage. As the damage is an irreversible non-decreasing variable, h is a smooth positive function and can be integrated:

$$d = \tilde{d}(\alpha)$$
 with $h(\alpha) = \frac{\mathrm{d}\tilde{d}(\alpha)}{\mathrm{d}\alpha}$ and $\tilde{d}(0) = 0$ (10)

Finally, the stress-strain relation of damaged hyperelastic material is given by Equations 6 and 10.

3.2 Similarity between CDM and two-networks (phases) models

The last problem to solve is the choice of the measure of deformation state α .

In the general theory of thermodynamics of irreversible processes, the equation of state for an internal variable has to be written in terms of the thermodynamical force associated with this variable. In the present case, it means that the measure $\alpha(t)$ should be chosen as a function of $-W_0$. The first authors who proposed to applied CDM to the Mullins effect adopted this approach (Simo 1987, De Souza Neto et al. 1984, and Miehe 1995). It yields to:

$$\alpha = f(W_0) \tag{11}$$

Even if *f* has not to be a norm, it can be reasonably stated that *f* is a positive increasing function on $[0, +\infty)$ and f(0)=0.

This choice exhibits one major problem: the form of the strain energy function should be postulated before derivation of the whole model. Moreover, due to physical considerations, it seems to be more appropriate to formulate explicitly the damage evolution equation in terms of the deformation state. That was proposed by Govindjee & Simo (1991, 1992), and later by Miehe & Keck (2000). It is to note that this approach can be related to the application of the two-network or -phase theories to stress-softening in rubber. Indeed, authors consider that the Mullins effect is due to a change in the microstructure of the network. The evolution of the network is described by a network reformation function that depends on the maximum strain state endured by the material in its life. The adopted deformation state is measured through a scalar function, which depends on I_1 and I_2 . As proposed by Beatty & Krishnaswamy (2000), this function can be seen as a damage parameter (see footnote 5 in their paper). A practical study of the choice of the driving damage parameter, the maximum stretch or the maximum virgin strain energy, was recently proposed by Laiarinandrasana et al. (2001).

Here, this second approach is adopted and the scalar measure of the deformation state is written as:

$$\alpha = g(I_1, I_2) \tag{12}$$

Using the properties of f, the material isotropy, the convexity of W_0 and the chain rule differentiation, it can be established that (Chagnon et al. submitted):

- *g* is defined on $[3,+\infty)\times[3,+\infty)$,
- g is differentiable in both I_1 and I_2 on $[3, +\infty) \times [3, +\infty)$,
- $\partial g/\partial I_1 > 0 \text{ and } \partial g/\partial I_2 \ge 0,$
- the choice $g(I_1=3,I_2=3)=0$, that ensures that the measure is null for the virgin unloaded state, is recommended.

Some applications of these developments are presented in the next section.

4 APPLICATIONS

4.1 Identification of the constitutive equation

The material used for the experimental part of this work is a carbon black filled natural rubber. The percentage of fillers is about 30%.

In order to determine material parameters for the virgin strain energy function W_0 and the damage equation of state 10, both cyclic uniaxial tensile and simple shear tests were performed. The uniaxial tensile experiments were conducted on flat coupon specimens and the simple shear specimens are four blocks samples. In order to simplify the fitting task, results of simple shear experiments were transformed into pure shear data. All experiments were performed under controlled strain conditions. For several strain levels, specimens underwent five cycles at a constant strain rate.

As in most of the studies concerned with the Mullins effect, the present paper only deals with the loss of stiffness of the material. Therefore, the stress-softening behaviour should be separated from other inelastic phenomena such as hysteresis and relaxation. The details of the correction method adopted are given in Marckmann et al. (2002) and the corrected experimental data for uniaxial tensile experiments are presented in Figure 1. The pure shear results are similar.



Figure 1. Corrected data for cyclic uniaxial tensile experiments.

In the present study, the emphasis is laid on the stress-softening phenomenon, so that a simple phenomenological strain energy function is chosen. In order to satisfactorily reproduce the large strain response of the material, the Yeoh model is adopted (Yeoh 1993). The corresponding strain energy function is given by:

$$W = C_{10}(I_1 - 3) + C_{20}(I_1 - 3)^2 + C_{30}(I_1 - 3)^3$$
(13)

where C_{10} , C_{20} and C_{30} are the three material parameters. As this model only depends on the first principal stretch invariant, it is not well-adapted to moderate strain. Nevertheless, its mathematical simplicity and ability to reproduce the whole behaviour of elastomers are sufficient qualities for the present study. Once the virgin strain energy function is chosen, the form of the damage equation of state has to be established. As detailed in the theoretical part of the paper, the present approach focuses on the use of a measure of deformation expressed as a function of the two first principal invariants of the stretch tensor **B**. As a first approach, we adopt a very simple measure of the deformation state that only depends on I_1 :

$$\alpha = \sqrt{I_1/3 - 1} \tag{14}$$

This measure is relevant with regard to the properties exhibited in the theoretical section.

The method adopted to determine the material parameters being thoroughly examined elsewhere (Chagnon 2001), only principal results are given. First, in order to establish the form of the damage function, the properties of the damage formulation are used: on secondary loading curves, the damage remains constant and the stress ratio between two secondary loading curves does not depend on the deformation level. As a consequence, we show that the damage evolution equation can be fitted by an exponential-like decreasing function, in both uniaxial tensile and pure shear cases (Chagnon, submitted):

$$\widetilde{d}(\alpha) = d_{\infty} \left[1 - \exp\left(-\frac{\alpha_m}{\eta}\right) \right]$$
(15)

where d_{∞} and η are material parameters.

Five material parameters have to be determined: three for the virgin strain energy, i.e. C_{10} , C_{20} and C_{30} , and two for the damage equation of state, i.e. d_{∞} and η . The identification results are compared with experiments in Figures 2 and 3 for uniaxial tensile and pure shear deformation states, respectively.



Figure 2. Identification for uniaxial tensile results: (\cdots) experiments, (--) our model.



First principal stretch Figure 3. Identification for pure shear tensile results: (\cdots) experiments, (--) our model.

Theoretical results are globally in good agreement with experiments; so, the model describes successfully the transition between different loading curves in cyclic experiments and exhibits the of the maximum strain influence on the accommodated behaviour. However, the present model is not able to satisfactorily predict the behaviour of the material at the intersection of secondary loading curves with the primary curve. Indeed, the high curvature of secondary loading due to strain-hardening is not well curves reproduced. Nevertheless, such results with only five material parameters are quiet good, and the use of the present model in finite element simulations will provide quality results.

4.2 Numerical results

Here, we choose to implement the present constitutive equation in Abaqus by using the UMAT facility.

The example considered is a well-known plane stress hyperelasticity problem where an initial $20 \times 20 \text{ mm}^2$ strip with a circular hole of diameter 10 mm is stretched in both horizontal and vertical directions. The corresponding initial mesh is shown in Figure 4(a). This example exhibits the strong dependence of the accommodation on the deformation history. Three different deformation paths are investigated (amplitudes of cycles are identical, namely equal to 185%) and their corresponding damage distributions are presented in Figure 4(b), (c) and (d).

The influence of the loading history on the damage distribution can be highlighted by comparing results in Figure 4(c) and 4(d). The two samples were subjected to the same deformation state, said 185% in both directions 1 and 2. Only loading paths differ. In the case 4(c), the sample is first stretched then relaxed in direction 2. This intermediate state corresponds with Figure 4(b). This first cycle weakens the structure in the neighbourhood of the hole, in direction 1. Then, the second cycle applied in direction 1 modifies a new unloaded non-homogeneous damage distribution. So, the final damage distribution is non-symmetric. In the loading case 4(d), both cycles (in directions 1 and 2) are applied simultaneously and the final damage distribution respects the structural symmetry.



Figure 4. Finite element simulation of a square perforated sheet: (a) undeformed mesh, (b) damage distribution after a cycle in dir. 2, (c) damage distribution after a cycle in dir. 2 followed by a cycle in dir. 1, (d) damage distribution after a cycle in the diagonal direction.

5 CONCLUDING REMARKS

Connections between the two phenomenological theories used for the Mullins effect, i.e. damage mechanics and two-network (or two-phase) approaches, are now established. Then, the only remaining difficulty is the determination of the equation of state that drives the parameter used to correct the strain-energy function: the damage the network reformation parameter, or transformation function between hard and soft regions of rubber, depending on the terminology adopted. Here, we showed that the use of the maximum deformation endured previously by the material as the damage criterion is sufficient to reproduce the global behaviour of elastomers subjected to the Mullins effect, but not enough accurate to describe strain-hardening. Thus, the damage equation of state should be also written in terms of the current deformation measure and not only in terms of the maximum deformation measure. Such approaches were recently proposed by Miehe & Keck (2000) and Elias-Zuniga & Beatty (2002).

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