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Comparison of recent models for the Mullins effect: formulation and finite element implementation

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ABSTRACT: The present paper focuses on the modelling of the Mullins effect in elastomers. Different approaches respectively based on physics, damage mechanics and the two-network theory are presented. The pros and the cons to their formulation and finite element implementation are discussed. It appears that all these approaches are complementary and present different limitations due to their formulations.

1 INTRODUCTION

Elastomers present a loss of stiffness after the first loading cycle of a fatigue experiment (Mullins, 1969). It has been proved that this phenomenon is only dependent on the maximum deformation previously reached in the history of the material. It is quite important to model it because the mechanical behaviour of rubber products is highly modified by this softening phenomenon. Moreover, as the Mullins effect depends on the maximum deformation endured previously, material points of the product are not identically affected. As a consequence, it is not acceptable to determine experimentally an accommodated hyperelastic constitutive equation for the material; then stress-softening should be explicitly included in the model.

The present paper only focuses on the Mullins effect, others phenomena exhibited by elastomers, such as creep and hysteresis, are not taken into account. The behaviour is then considered time-independent and can be schematically represented by Figure 1 that corresponds to a tensile cyclic test. The virgin undamaged material is first stretched as the extension ratio reaches λ_I and the stress follows the path I. Then the unloading from λ_I to 0 follows the path I'. The second loading from 0 to $\lambda_{II} > \lambda_I$ first follows the path I' until $\lambda = \lambda_I$ then it follows the path II. The second unloading from stretch ratio λ_{II} to 0 follows the path II' which is different than the path I'. At a given stretch, the stress on II' is lower than the stress on I'. Repeating this process, the loading path corresponding to the increase of stretch from 0 to λ_{III} is the path that joins II' and the part III of the virgin curve. Finally, the corresponding unloading follows the path III'.

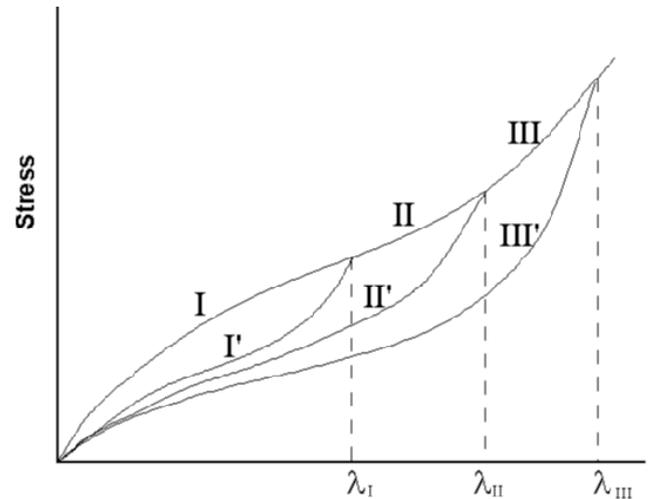


Figure 1. Schematic behaviour of a hyperelastic material with stress-softening.

Different constitutive equations for the Mullins effect will be presented and compared using both uniaxial analytical results and finite element simulations. Limitations of each model will be highlighted.

2 MODELS FOR THE MULLINS EFFECT

Different approaches have been used for many years to simulate the stress-softening phenomenon in elastomers: the physical approach that attempts to describe the evolution of the polymer network under deformation, the phenomenological two-phase network theory which considers that the material is constituted of a soft and a hard phases, and the continuum damage mechanics that assimilates stress-softening to damage.

Bueche (1960,1961) developed an uniaxial model by assuming that the Mullins effect is due to the breakdown of links between filler particles and

chains, and that it depends on the maximum stretch history. Harwood *et al.* (1966) suggested that the softening of the material occurs entirely in the rubber matrix, because stress-softening is also observed in unfilled rubbers. Nevertheless, no efficient, i.e. precise and numerically simple, physical-based constitutive equation exists. Recently, Marckmann *et al.* (2002) proposed a new constitutive equation (denoted M model through the rest of the paper) that describes the evolution of the network considering the rupture of links between polymer chains. The increase of chain length and the decrease of the number of chains reflect this evolution by volume unit as functions of the maximum deformation previously endured by the material. This approach is introduced in the eight-chain model of Arruda and Boyce (1993):

$$W = C_r N \left[\frac{\lambda_{chain}}{\sqrt{N}} \beta + \ln \frac{\beta}{\sinh \beta} \right] \quad (1)$$

with

$$\beta = \mathcal{L}^{-1} \left[\lambda_{chain} / \sqrt{N} \right] \quad (2)$$

where \mathcal{L} is the Langevin function. C_r and N are the material parameters which respectively represents the density of chains per unit of volume and the number of monomers per chain. To describe stress-softening, material parameters depends on the maximum deformation. Using experimental results, authors show that the evolution of material parameters can be driven by exponential functions: a decreasing one for the number of chains per unit of volume and an increasing one for the number of monomers per chains.

Mullins and Tobin (1957) have proposed a model based on the two-phase theory. They consider that the material contains soft and hard rubber phases. Under loading, hard rubber is transformed into soft rubber. The evolution of the ratio between phases is supposed to depend on the maximum deformation. Johnson and Beatty (1993) used this approach to model uniaxial tensile tests, by using an accommodation function to reproduce loss of stiffness. The form of this function evolved during years and the last proposal is due to Zuñiga and Beatty (2002) who proposed the following strain energy function W (denoted ZB model in the following):

$$W = F(m, M) W_0 \quad (3)$$

where W_0 is a classical hyperelastic strain energy function and the accommodation function F is given by:

$$F(m, M) = \exp(-b\sqrt{M-m}) \quad (4)$$

where b is a material parameter, and m and M represent measures of deformation, expressed thanks to the strain invariants :

$$m = \sqrt{I_1^2 - 2I_2} \quad \text{and} \quad M = \max_{0 \leq s \leq t} m(s) \quad (5)$$

Stress-softening is here described by the difference between the current state and the maximum deformed state previously endured by the material.

The continuum damage mechanics has often been used to model the Mullins effect even if this phenomenon is not a strictly speaking damage phenomenon. For example, it can be recovered with time and annealing accelerates this recovery. A thermodynamic variable D is introduced to represent stress-softening. The general theory of damage mechanics was introduced by Lemaitre and Chaboche (1990). As applied to hyperelasticity, it yields to:

$$W = (1 - D) W_0 \quad (6)$$

Recently, Chagnon *et al.* (2003, *subm.*) developed a damage mechanics approach (denoted C model in the following) and established the evolution equation of the damage variable thanks to second loading curves. It is expressed thanks to the first strain invariant and presents an exponential form:

$$D = D_\infty \left(1 - \exp\left(-\frac{I_1^{\max}}{\eta}\right) \right) \quad (7)$$

where D_∞ and η are material parameters. This model is similar to the discontinuous damage part of the constitutive equation proposed by Miehe (1995). The major difference is the choice of the damage criterion. Here this criterion is given as a function of the deformation state whereas Miehe used the strain energy density as a measure.

The three models described above, were developed using different approaches. They are now compared in the two next sections of the paper.

3 SIMPLE LOADING SOLUTIONS

The three previous models are studied using experimental data that correspond with a carbon-black filled rubber. Their material parameters will be determined by fitting uniaxial tensile and pure shear experiments.

First, it is to note that there is a major difference between the two-phase approach (ZB model) and the two other ones. The M and C models accumulate softening during the loading process, and the ZB stress-softening function evolves during unloading. This approach simplifies the identification task, because the two parts of the model, the hyperelastic strain energy and the stress-softening function, are independent and can be fitted separately. Nevertheless, it is difficult to explain physically the evolution of the stress-softening function during the unloading part of cycles.

The C and ZB models impose the choice of a strain energy function because it is not explicitly incorporated in the models. It is important to choose a density that can describe the whole behaviour of the material (small and large strain) but with few parameters (to simplify the identification task).

For the ZB model, every strain energy functions can be chosen, due to the formulation simplicity emphasised above. The model proposed by Hart-Smith (1966) has only three parameters and is able to describe the whole behaviour:

$$W = C_1 \int \exp(C_3(I_1 - 3)^2) dI_1 + C_2(I_2 - 3) \quad (8)$$

The first term of Equation 8 describes the whole behaviour of the rubber, and the second term improves the results at moderate strain. Thus, it can be omitted to simulate the whole behaviour. A simulation of uniaxial tests is presented in Figures 2 and 3.

The problem is quite different for the C model as the damage variable evolves during the loading part

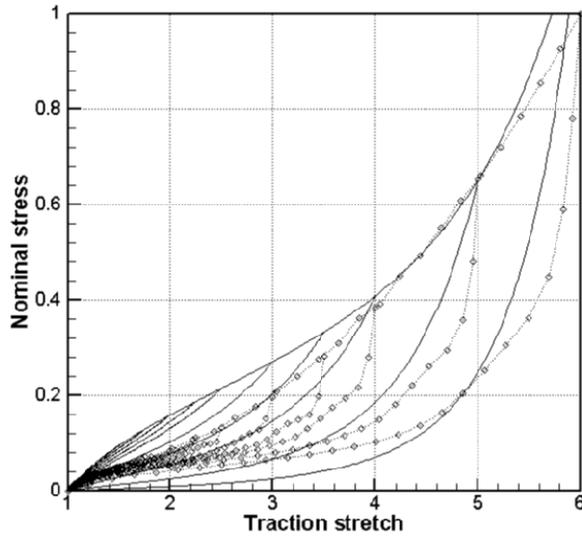


Figure 2. Uniaxial tensile results: (—) ZB model, (...) experimental data.

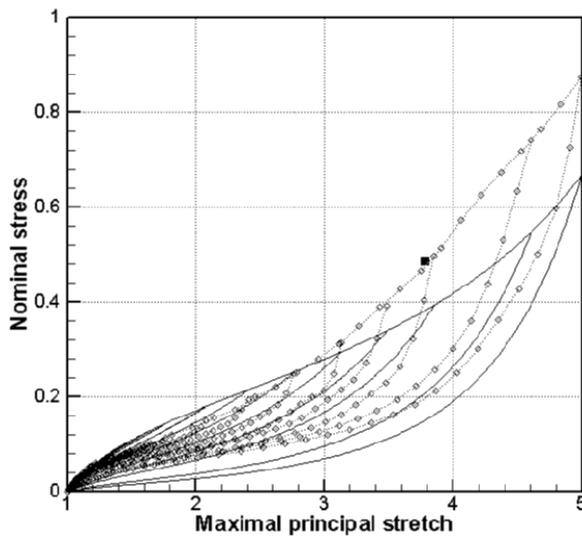


Figure 3. Pure shear results: (—) ZB model, (...) experimental data.

of the first cycle. Then, the first loading curve is described by both the hyperelastic behaviour and the damage function. It is important to choose a strain energy function that is compatible with the damage evolution law. Energy densities with important hardening cause abnormal curvatures of the model; then, a regular form of W_0 must be selected. The energy density proposed by Yeoh (1990) is chosen:

$$W_0 = C_{10}(I_1 - 3) + C_{20}(I_1 - 3)^2 + C_{30}(I_1 - 3)^3 \quad (8)$$

The corresponding simple results are presented in Figures 4 and 5.

The M model basically includes a given strain energy function. Due to network changes, material parameters evolve during the loading part of the first cycle. As a consequence, this first loading curve cannot be fitted independently. In fact, the evolution of material parameters of this model with deformation leads to a better simulation of the first loading curve (Meissner, 2000). Simulation results obtained with the M model are presented in Figures 6 and 7.

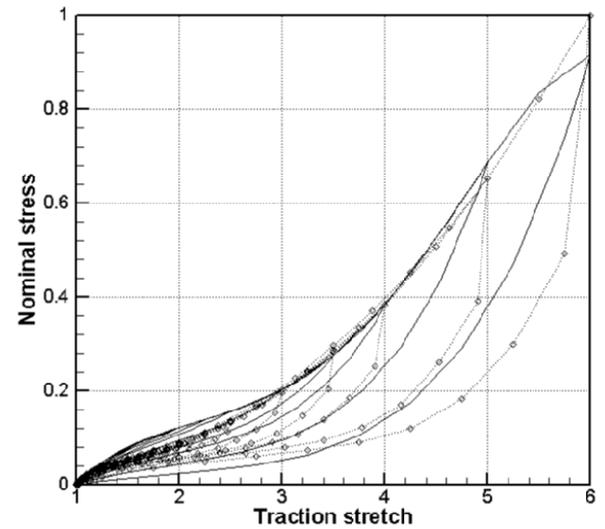


Figure 4. Uniaxial tensile results: (—) C model, (...) experimental data.

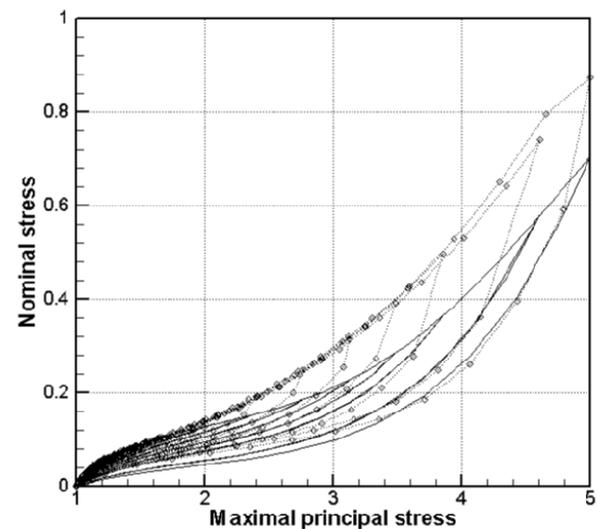


Figure 5. Pure shear results: (—) C model, (...) experimental data.

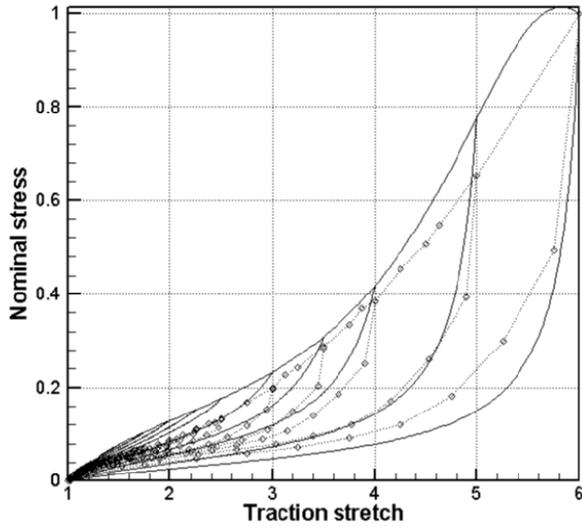


Figure 6. Uniaxial tensile results: (—) M model, (...) experimental data.

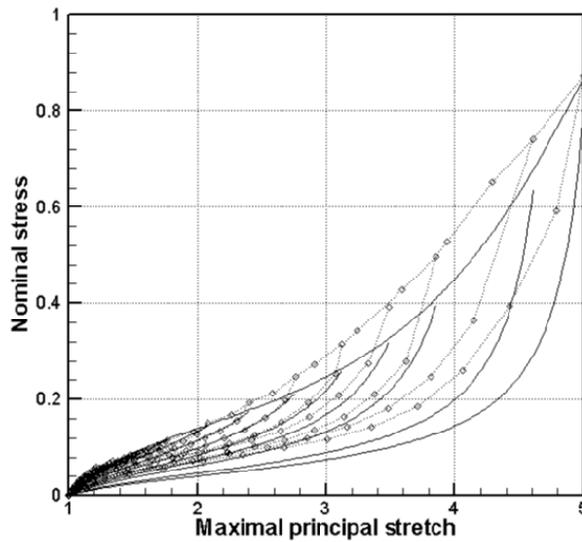


Figure 7. Pure shear results: (—) M model, (...) experimental data.

Figures 2 to 6 show that models have different characteristics. Due to its multiplicative form, the C model second loading curves are proportional. This makes difficult the description of the curvature of the second loading paths. In fact, this kind of modelling cannot take into account the strain-hardening phenomenon that takes place as secondary curves intersect the first loading curve. Second loading curves corresponding with the C model are too smooth. The two others models do not present this difficulty. The M model being based on the eight-chain model hyperelastic constitutive equation, it simulates correctly the strain-hardening. The ZB approach (associated with the Hart-Smith strain energy function) can satisfactorily describe strain-hardening thanks to the exponential function. However, it appears that the initial stiffness of loading curves decreases as the strain-hardening is more important. As a consequence, a good description of hardening reduces too

importantly the initial slope of second loading curves.

The M model is revealed to be the most efficient to describe the Mullins effect. Nevertheless, it is limited by its formulation: it is written in terms of principal strain instead of strain invariants. The fifth order Taylor development of the eight-chains model can be used to overcome this difficulty, but it is not well-adapted to the description of the Mullins effect, the use of the Langevin function being fundamental in the approach proposed by Marckmann et al. (2002).

4 NUMERICAL IMPLEMENTATION

The three previous constitutive equations were implemented in the finite element software Abaqus, thanks to the UMAT facility. The implementation necessitates the computation of both Eulerian stresses and the updated Lagrangian elasticity tensor.

The Arruda-Boyce eight-chain model is already implemented in Abaqus, under its Taylor development of the first strain invariant. This avoids difficulties induced by the use of principal strains instead of strain invariants. Nevertheless, as shown above, the M model necessitates the use of the original eight-chain model, because the strain-hardening must be well-described. The inverse of the Langevin function generates numerical instabilities and convergence difficulties at large strain. Finally, this model is very efficient in for analytical problems under simple strain states, but it cannot be used in finite element applications.

The C and ZB models being written in terms of strain invariants, their numerical implementations are easy and the convergence is easier to ensure. Due to its form, the C model can be used for very large range of calculations. The ZB model exhibits some difficulties mainly due to the choice of the strain measure (Equation 5). The square function leads to the occurrence of a vertical tangent. It is similar to the difficulties evoked above for the Langevin function. A simple method to overcome this difficulty is the change of the strain measure.

Results qualities of these models are similar to those obtained for simple problems. Let us recall that the M and ZB models cannot converge for very large strain. In the convergence range of the three models, they are able to describe the local loss of stiffness of the material and the non-homogeneity of the structure after a first stretching. An example is given in Figure 8 for an engine mount submitted to compressive loading. Results are given for the C model. It appears that the stress-softening level is quite different in the part, i.e. some zones are not damaged

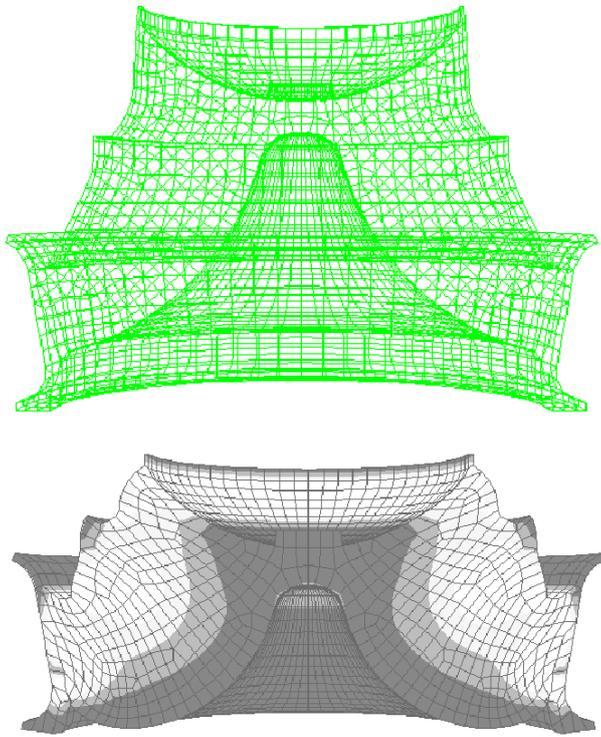


Figure 8. Damage level in an engine mount after a compressive loading (maximum damage zone : 25%).

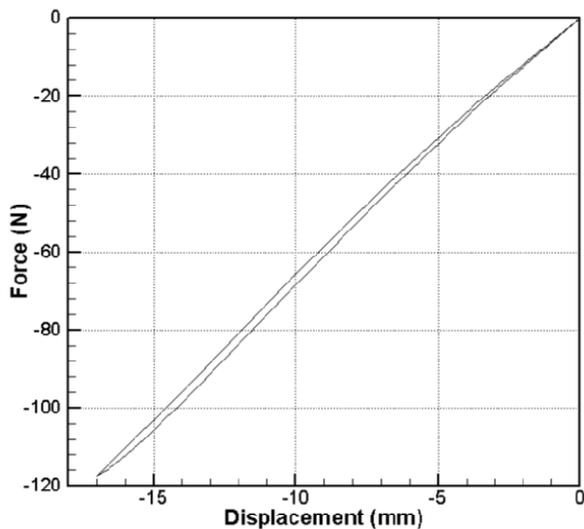


Figure 9. Force-displacement response of the engine mount.

and others zones are 25% damaged. However, the global response of the mount was not changed after this loading (see Figure 9). This example highlights the importance of considering the Mullins effect in the constitutive equation: even it is overall unimportant, it is fundamental to consider it in high-stressed parts of structures.

5 CONCLUSION

The three models studied in this paper have both advantages and limitations. For simple loading problems, the M model is the more efficient since it permits correct description of the form of the second

loading curves, especially strain-hardening. However, in order to use a finite element code, formulations in terms of strain invariants with regular functions leads to a better convergence. In this way, even if the damage C model exhibits worse results for simple problems than the two other models, its formulation is well-adapted to numerical applications. At our opinion, the ZB model could give such good results by using another strain measure. Its advantage compared with the C model is its ability to describe an important loss of stiffness. Then, it can be used to evaluate the energy dissipated between the two first loading cycles, but it cannot satisfactorily describe initial slopes of secondary stress-strain curves.

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