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### Continuum mechanical model for the Mullins effect in swollen rubber

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**Abstract.** The present paper deals with the continuum mechanical modeling of Mullins effect observed in swollen rubber under cyclic loading conditions. For this purpose, the concept of Continuum Damage Mechanics (CDM) applied to rubber materials is adopted and extended in order to take into account the swelling level. The damage exhibited by the material due to Mullins effect is assumed to be isotropic and is described by a scalar damage parameter which depends on the swelling level and on the maximum deformation experienced by the material during loading history. Results show that the proposed model is qualitatively in good agreement with experimental observations.

#### Introduction

Rubbers are widely used in engineering application such as vibration isolator, seals, gaskets, pipe, etc. During the service, they are subjecting to fluctuating mechanical loading which can lead to fatigue failure. Moreover, in engineering applications involving the contamination of aggressive liquids into the rubber components, swelling becomes a major issue since it decreases significantly the mechanical properties of rubber [1]. Thus, the understanding of the mechanical response of swollen rubber is essential for durability analysis of such components.

In the present paper, the continuum mechanical modeling of Mullins effect in swollen rubbers under cyclic loading conditions is addressed. More precisely, the concept of Continuum Damage Mechanics is adopted and extended in order to account for swelling level. For this purpose, the mechanical testing probing the Mullins effect in swollen rubber is conducted. The data is subsequently used to construct the damage function describing Mullins effect.

#### **Continuum Mechanical Framework**

**Experimental Observation.** The stress-strain responses of dry and swollen rubbers under cyclic loading at different maximum strains are presented in Fig. 1. In this figure, the secondary curves correspond to the reloading paths. The stress level during primary curve becomes lower as the swelling level increases. Moreover, as in dry rubber, it is observed that the swollen rubber also exhibits Mullins effect. However, their amount depends on the swelling level [2].

**Constitutive Equation.** At a continuum scale, swollen rubber is considered as homogeneous, isotropic and hyperelastic. Moreover, it is subjected to isotropic damage due to Mullins effect [3]. Since the volume of swollen rubber does not significantly change during the deformation, the material is assumed to be incompressible. Thus, the mechanical response of the swollen rubber can be entirely characterized by the existence of a strain energy function  $W_S$  which depends on the two invariants  $I_1$  and  $I_2$  of the left Cauchy-Green strain tensor  $\underline{B}$  relative to the *swollen-unstrained* configuration, on



Fig. 1. Mullins effect observed in dry and swollen rubbers.

the scalar parameter *d* describing isotropic damage due to Mullins effect and on the swelling level  $J_s = V/V_0$  as follow:

$$W_{s} = W_{s}(I_{1}, I_{2}, d, J_{s})$$
<sup>(1)</sup>

Following the concept of Damage Mechanics by Lemaitre and Chaboche [4],  $W_S$  can be considered as the product of the surface reducing parameter 1 - d and the strain energy function of the hypothetically virgin undamaged (due to Mullins effect) swollen material  $W_{OS}$ 

$$W_{S} = (1 - d) \cdot W_{OS}(I_{1}, I_{2}, J_{s})$$
<sup>(2)</sup>

It is important to note that the damage that we address in the present work corresponds to the Mullins effect only. Considering the second law of thermodynamics, it can be shown that the Cauchy stress tensor in the incompressible swollen rubber is given by:

$$\underline{\underline{\sigma}} = -p\underline{\underline{I}} + (1-d) \cdot \left[ 2 \left( \frac{\partial W_{OS}}{\partial I_1} + I_1 \frac{\partial W_{OS}}{\partial I_2} \right) \underline{\underline{B}} - 2 \frac{\partial W_{OS}}{\partial I_2} \underline{\underline{B}}^2 \right]$$
(3)

where the scalar p is the Lagrange multiplier due to incompressibility assumptions and is to be determined from the equilibrium equations and boundary conditions. Classically, it is mainly recognized that the Mullins effect exclusively depends on the maximum deformation endured by the material during its history [5]. Therefore, a scalar measure of the deformation state and its maximum which depends on the deformation history should be defined. Simo [6] and Miehe [7] used the virgin

strain energy density while [3] adopted the elongation of polymeric chain issued from 8-chain model as the measure of deformation state. In the present study, following [3] and [7], we adopt the discontinuous exponential-like damage function to desribe Mullins effect. Moreover, the damage function is extended in order to account for the swelling level. For this purpose, the following phenomenological damage function is proposed:

$$d = d(\overline{\alpha}, J_s) = d_s(J_s) \cdot \left[ 1 - \exp\left(\frac{-\overline{\alpha}}{\eta_s(J_s)}\right) \right]$$
(4)

where:

$$d_{s} = d_{s}(J_{s}) = d_{OS} + d_{1}(J_{s} - 1)^{2} \qquad \text{and} \qquad \eta_{s} = \eta_{s}(J_{s}) = \eta_{OS} + \eta_{1}(J_{s} - 1) \qquad (5)$$

In the above equation,  $d_{OS}$ ,  $d_1$ ,  $\eta_{OS}$  and  $\eta_1$  are material parameters. In the case of dry rubber, i.e. when  $J_s=1$ ,  $d_{OS}$  and  $\eta_{OS}$  correspond to the dimensionless maximum (possible) damage and the damage saturation parameter respectively [8].  $\overline{\alpha}$  is the maximum value of deformation measure in the loading history. By choosing polymeric 8-chain elongation as the measure of deformation state, its maximum is defined by:

$$\overline{\alpha}(t) = \max_{\tau \in ]-\infty, t]} [\alpha(\tau)] \qquad \text{and} \qquad \alpha(t) = \sqrt{\frac{I_1(t)}{3}} - 1 \tag{6}$$

According to Treloar [9], swelling of rubber in liquid molecules is a purely physical mixing or interdiffusion process in which the two components may be regarded as chemically neutral. Consequently, from the standpoint of mechanical response, the only effect of the swelling is to reduce the modulus in inverse proportion of the swelling ratio. However, our previous investigation revealed a deviation from the Treloar's theory [10]. Thus, we propose the following generalized Treloar's theory which relates the hyperelastic strain energy functions of dry  $W_{OD}$  and swollen one  $W_{OS}$ :

$$W_{OS}(I_1, I_2, J_s) = (J_s)^{-n} \cdot W_{OD}(I_1, I_2)$$
(7)

where *n* is a constant.

#### **Results and Discussion**

**Special Case of Uniaxial Extension.** Without losing generality, we focus our attention on the case of uniaxial extension. Considering the general form of the stress-strain curve, it appears that the Ogden strain energy density function fits well [11]:

$$W_{OS}(\lambda_1, \lambda_2, \lambda_3, J_s) = (J_s)^{-n} \cdot \sum_{p=1}^{3} \frac{\mu_p}{\alpha_p} \left( \lambda_1^{\alpha_p} + \lambda_2^{\alpha_p} + \lambda_3^{\alpha_p} \right)$$
(8)

where  $\lambda_1$ ,  $\lambda_2$ ,  $\lambda_3$  are the three principal stretches while  $\mu_p$  and  $\alpha_p$  are material parameters. By considering the boundary conditions to determine the Lagrange multiplier *p*, the analytical expression for the first Piola-Kirchhoff stress (engineering stress) is:

$$P_{1} = (1-d) \cdot (J_{s})^{-n} \cdot \sum_{p=1}^{3} \mu_{p} \left( \lambda_{1}^{\alpha_{p}-1} - \lambda_{1}^{\frac{-\alpha_{p}-2}{2}} \right)$$
(9)

where d is given by Eqs. (4) and (5). By fitting simultaneously the experimental data, the following material parameters are obtained:



Fig. 2. Comparison between the model and experimental data.

**Comparison with Experimental Data.** Fig. 2 illustrates the comparison between the model and experimental data. In general, the model gives qualitatively good agreements with experimental data. The stress-softening is well described by the model for dry rubber and for swollen rubbers, in particularly at relatively low strain. More discrepancies are observed in swollen rubber for a larger strain level. Indeed, the model appears to overestimate the stress response. The effect of swelling is well described as shown by the primary curves where the model is capable to predict the general stress response of swollen rubbers.

The discrepancies observed between the model and experimental data in large strain range (where strong reinforcement due to strain hardening occurs) can be explained by the limitation of the model itself. Indeed, as highlighted in [3], this limitation is inherent in the choice of the damage mechanism: since the damage variable is written in terms of a maximum deformation state, theoretical secondary curves are proportional. This proportionality is not satisfied at large strain when the reinforcement becomes apparent.

#### Conclusions

In the present work, the modeling of Mullins effect (stress-softening) in swollen rubber under cyclic loading was addressed. In order to describe the damage in material resulting to stress-softening, an extension version of CDM model developed by Chagnon et al. [3] was considered. More precisely, the damage parameter is enriched by including the effect of swelling on the stress-softening. Results show that the model gives qualitatively good agreements with experimental data.

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