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# Multiphysics Coupling In Aging Process of Filled-Rubber

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***Abstract*: Filled rubbers are used popularly in damping parts which can be found in automobile sector or in building sector. However, the mechanical properties of material depend sensitively on temperature, chemical composition and environment conditions. In fact, the mechanical dissipation due to damping process leads to the increase of temperature considerably. Aging process can be activated sequentially by the heat which result in the change of damping properties during usage time. This paper presents a new behavior model that considers the simultaneous effects of temperature, mechanical loads on the behavior of materials along with the aging of materials. With the assumption of internal variables related to aging phenomenon and visco-plastic behavior, the model is built in the thermodynamical framework. A fully coupled finite element formulation is proposed to solve simultaneously thermo, chemical and mechanical phenomenon appeared in this material. An example illustrates the number of applicability of the model to predict the behavior of materials under the effect of cyclic loads in extremely working conditions.***

***Keywords* : aging process, constitutive behavior, finite element method, Multiphysics couplings.**

## I. INTRODUCTION

Rubber materials are widely used in aviation, automotive, construction and industrial applications. Mechanical parts of this material are used in load-bearing, damping or gaskets applications. These parts are usually subjected to static load and dynamic load in large deformation. The environment conditions can also affect on the use of the parts due to the aging process activated by temperature variation. In fact, the dissipation energy from dynamic loads leads to increase considerably the temperature inside of rubber parts. Recent studies [1], [2], [3], [4] show that the mechanical characteristics and damping capacity of the material are significantly changed during aging process. Therefore, the study of the behavior of this material during usage time becomes a necessary issue to have appropriate design to increase the service life of these devices. The behavior model of materials taking into account the change of mechanical properties of materials due to the aging process under the effect of corrosive and chemical environments without consideration of mechanical loading is presented in [5]. However, due to chemical process activated by thermal and mechanical load, a fully coupled thermo-chemical mechanical model for vulcanization process of rubber is proposed in [6,7]. In this paper, a fully coupled thermo-chemical-mechanical model to describe the behavior of filled rubber during thermal aging. In particular, this model considers the influence of mechanical load on the aging process which is activated by the heat build-up phenomenon inside the material. A viscoplastic behavior of Bingham is proposed to describe the filled rubber. The study also provides a multi-field formulation and an appropriate numerical model to solve this strong coupling problem.

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## II. THERMODYNAMICAL FRAMEWORK

The material model is built based on thermodynamic framework with the assumption of local state. Chemical changes and nonlinear mechanical behavior are represented by internal variables. Therefore, the behavior of materials is defined from state variables: deformation gradient $\boldsymbol{F}$(X, t), absolute temperature $\theta$(X, t) and set of internal variables. The degree of aging of the material is defined by the mass ratio of the aging product and the total mass of the material:

$$\xi(X,t)=\frac{m_{aging}(X,t)}{m_{total}(X)}\in[0,1] \tag{1}$$

### A. Kinematics

To consider the compressible behavior of rubber material, the total transformation gradient is decomposed into the mechanical, thermal and chemical components as follows:

$$\mathbf{F}=\bar{\mathbf{F}}(J^{1/3}\mathbf{1}) \text{ with } J=J_M J_T J_C \tag{2}$$

Total volume variation ($J$) composes of thermal expansion, chemical shrinkage and compressible mechanical deformation. The above deformations are expressed in the following form:

$$\begin{aligned} J_T &= 1+\alpha\ \theta-\theta_{ref} \\ J_C &= 1+\beta\, g\ \xi,\xi_{ref} \end{aligned} \tag{3}$$

With α, β are the coefficient of thermal expansion, chemical shrinkage due to aging; The mechanical transformation gradient is then decomposed into elastic and non-elastic transformations:

$$\bar{\mathbf{F}}_M=\bar{\mathbf{F}}_e\bar{\mathbf{F}}_a \tag{4}$$

### B. Clausius-Duhem inequality

From the first and the second thermodynamical principle, the Clausius-Duhem inequality of irreversible processes is expressed in the following form:

$$\Phi = \boldsymbol{\sigma} : \mathbf{D} - \rho\dot{\Psi} - \rho s\dot{\theta} - \mathbf{q}\frac{grad(\theta)}{\theta} \geq 0 \tag{5}$$

Where q is the heat flux, σ is the Cauchy stress, D is the deformation speed tensor, s is the entropy, $\psi$ is the HelmHoltz free energy function.
Variation of free energy function:

$$\dot{\Psi} = \frac{\partial\Psi}{\partial\bar{\mathbf{B}}} : \dot{\bar{\mathbf{B}}} + \frac{\partial\Psi}{\partial\bar{\mathbf{B}}_{\mathbf{e}}} : \dot{\bar{\mathbf{B}}}_{\mathbf{e}} + \frac{\partial\Psi}{\partial J}\dot{J} + \frac{\partial\Psi}{\partial\theta}\dot{\theta} + \frac{\partial\Psi}{\partial\xi}\dot{\xi} \tag{6}$$

$$\dot{\bar{\mathbf{B}}} = \mathbf{L}\bar{\mathbf{B}} + \bar{\mathbf{B}}\mathbf{L}^T - \frac{2}{3}(1:\mathbf{D})\bar{\mathbf{B}}$$

$$\dot{\bar{\mathbf{B}}}_{\mathbf{e}} = \mathbf{L}\bar{\mathbf{B}}_e + \bar{\mathbf{B}}_e\mathbf{L}^T - \frac{2}{3}(1:\mathbf{D})\bar{\mathbf{B}}_e - 2\bar{\mathbf{V}}_{\mathbf{e}}\bar{\mathbf{D}}_a^o\bar{\mathbf{V}}_{\mathbf{e}} \tag{7}$$

$$\dot{J} = J(\mathbf{1}:\mathbf{D})$$

Where L is the rate of deformation tensor.
Replace equations (6), (7) into (5), the constitutive equations of material are written as follows:

$$s = -\frac{\partial\Psi}{\partial\theta}$$
$$\boldsymbol{\sigma} = 2\rho\left(\bar{\mathbf{B}}\frac{\partial\Psi}{\partial\bar{\mathbf{B}}} + \bar{\mathbf{B}}_e\frac{\partial\Psi}{\partial\bar{\mathbf{B}}_e}\right)^D + \rho J\frac{\partial\Psi}{\partial J}\mathbf{1} \tag{8}$$

The total dissipation energy in equation (5) are decomposed into a mechanical part, a thermal part and a chemical part due to aging process :

$$\Phi = \Phi_M + \Phi_C + \Phi_T \geq 0$$
$$\begin{cases} \Phi_M = 2\rho\left(\bar{\mathbf{B}}_e\frac{\partial\Psi}{\partial\bar{\mathbf{B}}_e}\right)^D : \bar{\mathbf{D}}_a^o \geq 0 \\ \Phi_C = -\rho\frac{\partial\Psi}{\partial\xi}\dot{\xi} \geq 0 \\ \Phi_T = -\frac{1}{\theta}q\nabla\theta \geq 0 \end{cases} \tag{9}$$

### C. Viscoplastic behavior of filled rubber

Filled rubber material exhibit the viscoplastic behavior. Therefore, a model of Bingham is chosen to describe nonlinear mechanical of filled rubber. This Bingham model is then assembled in series with chemical part and thermal expansion part (Fig. 1).

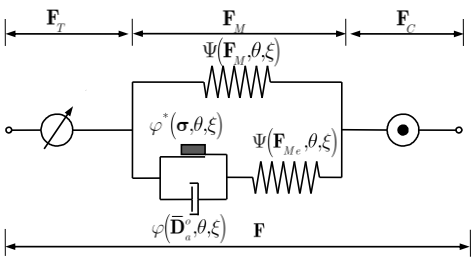


**Fig. 1 Viscoplastic behavior of Bingham model**

The behavior equations of material in equation (8) are deduced by the choice of the HelmHoltz free energy functions and pseudo-potential functions as follows:

$$\Psi = \psi_{eq}\ \bar{\mathbf{B}},\theta,\xi\ + \psi_{neq}\ \bar{\mathbf{B}}_e,\theta,\xi\ + \psi_{vol}\ J,\theta,\xi\ + \psi_T\ \theta\ + \psi_C\ \theta,\xi \tag{10}$$

$$\psi_{eq}\ \bar{\mathbf{B}},\theta,\xi\ = C_{10}\ \theta,\xi\ \ \bar{I}_1 - 3\ + C_{01}\ \theta,\xi\ \ \bar{I}_2 - 3$$
$$\psi_{neq}\ \bar{\mathbf{B}}_e,\theta,\xi\ = C_{10e}\ \theta,\xi\ \ \bar{I}_{1e} - 3$$
$$\psi_{vol}\ J,\theta,\xi\ = \frac{1}{2}K_V\ \ JJ_T^{-1}J_C^{-1} - 1\ ^2 \tag{11}$$
$$\psi_T\ \theta\ = C_0\left(\theta - \theta_{ref} - \theta\ Ln\left(\frac{\theta}{\theta_{ref}}\right)\right) - \frac{C_1}{\theta_{ref}}\frac{\theta - \theta_{ref}\ ^2}{2}$$
$$\psi_C = \left(k\ \theta\ \frac{1-\xi\ ^{n+1}}{n+1} + k\ \theta_{ref}\ \ 1 - \xi_{ref}\ ^n\ \xi\right) - \Psi_{Cref}$$

The function representing chemical reactions in equation (11) is only activated when the temperature reaches a certain limit according to Arrhenius law.
The pseudo - potential chosen for mechanical, thermal and chemical parts have the following forms:

$$\varphi^* = \varphi_M^*\ \sigma,\theta,\xi\ + \varphi_T^*\left(\frac{grad\ \theta}{\theta}\right) + \varphi_C^*\ A_C$$
$$\varphi_M^* = \frac{1}{2}\frac{< f(\boldsymbol{\sigma}_{neq},\theta,\xi) >^2}{\eta_{vp}\ \theta,\xi}$$
$$\varphi_C^* = \frac{1}{2\varpi\ \theta,\xi}\langle A_C\rangle^2 \tag{12}$$
$$\varphi_T^* = \frac{1}{2\ \theta\mathbf{K}_T\ \theta,\xi}\ grad\theta\ ^2$$
$$A_C = -\rho\frac{\partial\Psi}{\partial\xi}$$

Base on the choice of free energy and pseudo-potential above, we can deduce the stress strain relation and the evolution of internal variable correspond to the viscoplastic flow and to the degree of aging process:

$$\boldsymbol{\sigma} = \boldsymbol{\sigma}_{eq} + \boldsymbol{\sigma}_{neq} - p\mathbf{1}$$
$$\boldsymbol{\sigma}_{eq} = 2J^{-1}\left(\bar{\mathbf{B}}\frac{\partial\rho_0\psi_{eq}}{\partial\bar{\mathbf{B}}}\right)^D$$
$$\boldsymbol{\sigma}_{neq} = 2J^{-1}\left(\bar{\mathbf{B}}_e\frac{\partial\rho_0\psi_{neq}}{\partial\bar{\mathbf{B}}_e}\right)^D$$
$$p = -\frac{\partial\rho_0\psi_{vol}}{\partial J}$$
$$\dot{\bar{\mathbf{B}}}_e = \mathbf{L}\bar{\mathbf{B}}_e + \bar{\mathbf{B}}_e\mathbf{L}^T - \frac{2}{3}(\mathbf{1}:\mathbf{D})\bar{\mathbf{B}}_e - 2\frac{\langle f(\boldsymbol{\sigma}_{neq},\theta,\xi)\rangle}{\eta_{vp}(\theta,\xi)}\frac{\boldsymbol{\sigma}_{neq}}{\|\boldsymbol{\sigma}_{neq}\|}\bar{\mathbf{B}}_e$$
$$\dot{\xi} = \frac{1}{\varpi(\theta,\xi)}\langle A_C\rangle \tag{13}$$

## III. FINITE ELEMENT FORMULA OF MULTI-FIELD PROBLEM

### A. Balance equations

The equilibrium equations are represented in non-deformed configuration as follows:

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$$
\begin{aligned}
& DIV(\mathbf{\Pi}) + \rho_0 f_{vol} = 0 && \Omega_0 \\
& \mathbf{\Pi} N = F_{\text{surf}} && \partial\Omega_{0F} \\
& \mathrm{u} = \mathrm{u}_0 && \partial\Omega_{0\mathrm{u}} \\
& \rho_0 J^{-1} C\dot{\theta} = \Phi_M + \Phi_C + l_M + l_C + \rho_0 J^{-1} r - DIV(\mathbf{Q}) && (14) \\
& \mathbf{Q} N = \mathbf{Q}_0 && \partial\Omega_{0q} \\
& \theta = \theta_{ext} && \partial\Omega_{0T} \\
& \theta(t=0) = \theta_{\text{initial}} && \Omega_0
\end{aligned}
$$

Where $\Pi$ is Piola-Kirchoff stress, Q is the heat flux, r is the internal heat source, C is the specific heat capacity of the material.

The coupling terms in heat equation are latent heats of mechanical and chemical parts:

$$
l_M = \theta\left(\left(\frac{\partial \boldsymbol{\sigma}_{eq}}{\partial\theta} + \frac{\partial \boldsymbol{\sigma}_{neq,1}}{\partial\theta} + \frac{\partial \boldsymbol{\sigma}_{neq,2}}{\partial\theta}\right)^D - \frac{\partial p}{\partial\theta}\mathbf{1}\right) : \mathbf{D} - \theta\frac{\partial \boldsymbol{\sigma}_{neq}}{\partial\theta} : \bar{\mathbf{D}}_a^o
$$

$$
l_C = -\theta\frac{\partial A_C}{\partial\theta}\dot{\xi} \qquad (15)
$$

### B. Finite element formula

The multi-field finite element formula of the problem consists of displacement, temperature, chemical transformation, hydrostatic pressure related to the compressible, variation of pressure according to temperature, chemical transformation shown in formula (16).

$$
\begin{aligned}
& R_{\mathrm{u}}(\mathrm{u}, p, \theta, \xi, dp_\theta) = 0 \\
& R_p(\mathrm{u}, p, \theta, \xi, dp_\theta) = 0 \\
& R_\theta(\mathrm{u}, p, \theta, \xi, dp_\theta) = 0 \qquad (16) \\
& R_{dp_\theta}(\mathrm{u}, p, \theta, \xi, dp_\theta) = 0 \\
& R_\xi(\mathrm{u}, p, \theta, \xi, dp_\theta) = 0
\end{aligned}
$$

The interpolation function for displacement field, temperature field and chemical transformation field are quadratic. However, the linear interpolations are chosen for pressure and variation of pressure due to the numerical stability. The discrete formulas of the fields are written as follows:

$$
\begin{aligned}
& \mathrm{u}_h^e = \sum_{i=1}^{9} N_i^e U_i^e = \left[N_u^e\right]\left\{U^e\right\} \\
& \theta_h^e = \sum_{i=1}^{9} N_i^e \theta_i^e = \left[N_\theta^e\right]\left\{\theta^e\right\} \\
& \xi_h^e = \sum_{i=1}^{9} N_i^e \xi_i^e = \left[N_\xi^e\right]\left\{\xi^e\right\} \qquad (17) \\
& p_h^e = \sum_{i=1}^{3} \varphi_i^e p_i^e = \left[\Phi_p^e\right]\left\{p^e\right\} \\
& dp_{\theta h}^e = \sum_{i=1}^{3} \varphi_i^e dp_{\theta i}^e = \left[\Phi_{dp_\theta}^e\right]\left\{dp_\theta^{\,e}\right\}
\end{aligned}
$$

The stiffness matrix of multifield problem are deduced as follows:

$$
K\,\Delta d = r
$$

$$
K = \mathop{A}_{e}^{nel} K^e; \quad \Delta d = \mathop{A}_{e}^{nel} \Delta d^e; \quad r = \mathop{A}_{e}^{nel} r^e
$$

$$
K^e = \begin{bmatrix}
K_{\mathrm{uu}}^e & K_{\mathrm{u}p}^e & K_{\mathrm{u}\theta}^e & 0 & K_{\mathrm{u}\xi}^e \\
K_{p\mathrm{u}}^e & K_{pp}^e & K_{p\theta}^e & 0 & K_{p\xi}^e \\
K_{\theta\mathrm{u}}^e & 0 & K_{\theta\theta}^e & K_{\theta dp_\theta}^e & K_{\theta\xi}^e \\
K_{dp_\theta\mathrm{u}}^e & 0 & K_{dp_\theta\theta}^e & K_{dp_\theta dp_\theta}^e & K_{dp_\theta\xi}^e \\
K_{\xi\mathrm{u}}^e & K_{\xi p}^e & K_{\xi\theta}^e & 0 & K_{\xi\xi}^e
\end{bmatrix} \qquad (18)
$$

$$
r^e = \begin{Bmatrix} R_{\mathrm{u}}^e \\ R_p^e \\ R_\theta^e \\ R_{dp_\theta}^e \\ R_\xi^e \end{Bmatrix} \qquad \Delta d^e = \begin{Bmatrix} \Delta U^e \\ \Delta p^e \\ \Delta\theta^e \\ \Delta dp_\theta^{\,e} \\ \Delta\xi^e \end{Bmatrix}
$$

This formulation is then implemented in house finite element code to solve coupled problem.

## IV. RESULTS OF NUMERICAL SIMULATION

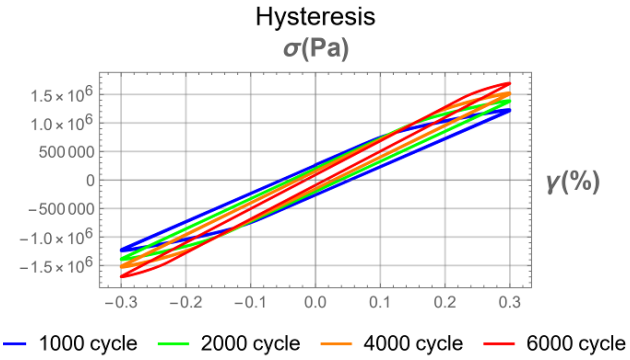


**Fig. 2: Nonlinear behavior of material during aging**

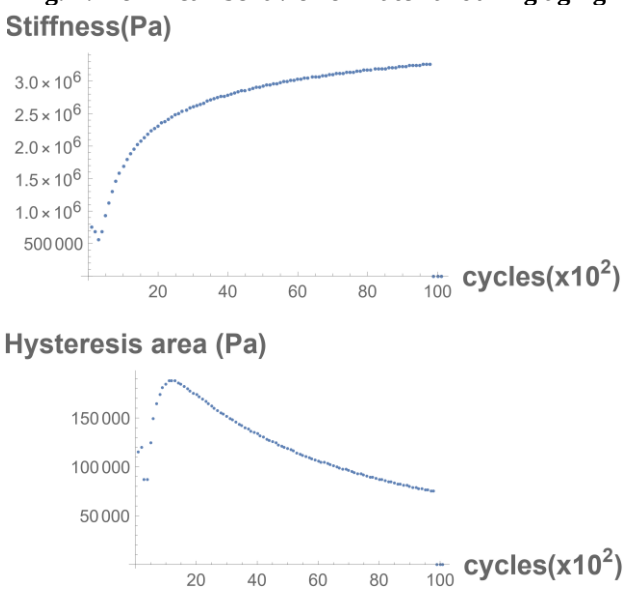


**Fig. 3: Evolution of damping capacity with aging time**

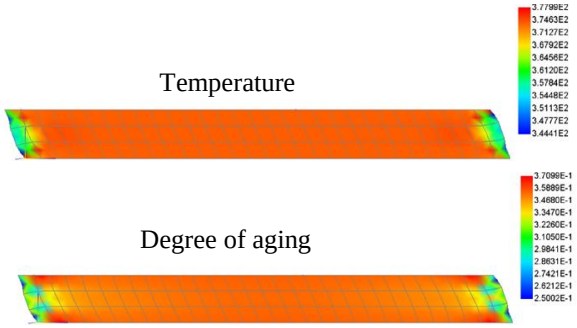


**Fig. 4: Distribution of temperature and degree of aging in rubber part**

An example of numerical simulation illustrates the strong interactions of thermo-mechanical-chemical fields in the aging process which is occurred when damping part subjected to dynamic loads with large amplitudes. We consider a rectangular rubber block subjected to cyclic shear load in large deformation of 50% at frequency of 3Hz. This extreme mechanical condition allows the heat generated by mechanical nonlinear process to activate the thermal aging. Assuming the process is adiabatic, there is no heat exchange with the outside. Heat generated only changes the temperature inside the sample. Initially, the aging level of the material is 0.2 and the initial temperature of the sample is 293K. Fig. 2 illustrates the change in stiffness of the material as well as the change of the hysteresis area in the aging process. The hysteresis shows that the material has a non-linear behavior of viscoplasticity. The aging process is activated by the temperature accumulated inside the material, then the aging process changes the mechanical properties of the material. The result also show that aging process decrease the capacity of damping of rubber material [1] ( see Fig. 3). Furthermore, the degree of aging in rubber part depend on the distribution of temperature generated in heat build-up phenomenon.

## V. CONCLUSION

In this paper, the author has proposed a model of thermo-chemical-mechanical behavior of rubber materials with mechanical behavior of visco-plastic in the rheology model of Bingham. This model is based on the thermodynamical framework of irreversible processes. To ensure the strong couplings of the fields, the finite element formula and the appropriate numerical method were developed to solve the thermal mechanical chemical problems simultaneously. The numerical example illustrates that the model can use to represent chemical changes of the aging process under the activation of temperature due to periodic mechanical loads. In addition, the model also shows the aging process of materials affected by mechanical loads. The changing of damping capacity during mechanical loading is considered to design absorber parts.

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