Modelling of Visco-hyperelastic behaviour of Carbon Black Filled Rubber

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Abstract
The mechanical behaviour of rubber-like materials is known to be time-dependent and to exhibit hysteresis upon loading.

In this work, the finite deformation, time-dependent behaviour of carbon black filled natural rubber has been studied experimentally by means of uniaxial tensile tests, multi-step relaxation and cyclic loading tests at room temperature. A visco-hyperelastic material model was incorporated into finite element package ABAQUS, aiming to capture the uniaxial large deformation time dependent response at room temperature. The model was used to simulate the material’s finite strain uniaxial response such as stress relaxation and hysteresis. The simulation results obtained were compared with the experimental data. The model has shown some success in describing finite deformation, time-dependent behaviour of a carbon black filled rubber but it under predict hysteresis behaviour.

1. Introduction
The use of rubbers especially carbon black filled rubbers has been widely seen in engineering applications. Typical examples of this include tires and engine mounts. However, the design of these rubber components has traditionally been driven by a process of trial and error. Recently computer-based techniques, such as finite-element analysis, are popularly used in the design of rubber components. Nevertheless, without accurate description of mechanical properties or constitutive equations to accompany them, simulation results can be erroneous and untrustworthy.

Rubber materials are known to have ability to undergo large deformation and fully recover to its original shape after load removal. They also exhibit a mechanical response which strongly depends on time, temperature and loading history. When rubber is deformed at a constant strain, the stress response decreases with time. This phenomenon is known as stress relaxation. Under cyclic loading, rubber dissipates energy due to hysteresis effects [1]. It has been known for many years that adding small amounts of filler particles, such as carbon black, to natural rubber can significantly improve both the stiffness and the strength of the material. However, time dependent and hysteresis are increased with the filler content [2].

In this work, the finite deformation, time-dependent behaviour of carbon black filled rubber has been studied experimentally by means of uniaxial tensile tests, stress relaxation and cyclic loading tests in an isothermal condition at room temperature. A visco-hyperelastic material model was derived aiming to capture the uniaxial large deformation, time dependent response at room temperature. The model was then incorporated into FE package ABAQUS and used to simulate response of carbon black filled natural rubber during cyclic loading condition. The simulation results obtained were compared with the experimental data to verify applicability of the material model.

2. Experimental Testing
In this work the mechanical properties of a natural rubber filled with 5% volume carbon black and an unfilled natural rubber were examined. The specimen geometry employed in these tests was taken from the ASTM 412 standard for the uniaxial tensile testing of elastomeric materials. Tests were performed at room temperature on an Instron universal testing machine model 5567.
An Instron non-contacting video extensometer was used for providing strain measurement on the narrow section of each test piece with a gauge length of 25 mm marked on the surface. The specimen was subjected up to 10 cycles of uniaxial tensile cyclic loading to two strain levels; 26 % and 62%, using a constant crosshead speed of 300 mm/min. During the first few load cycles
on a new carbon black filled specimen the material stiffness decreases [1]. The initial stress-induced softening is known as the Mullins effect. After a few load cycles the material’s response becomes stable, and the Mullins effect was removed permitting the hysteretic behaviour to be isolated from the Mullins effect. The stabilised stress-strain response (at 10th cycle) of the carbon black filled rubber was plotted together with the response of the unfilled natural rubber specimen for comparison in Figure 1. It can be seen from the figure that the unfilled natural rubber shows lower stiffness and smaller hysteresis loop (enclosed area of the stress-strain curve) comparing to the carbon black filled rubber.

To examine time dependent behaviour, after cyclic loading, uniaxial tensile stress relaxation were performed at a constant strain level of 45% up to 10 minutes. The plot of stress response over holding time is shown in Figure 2. It can be seen from the figure that the carbon blacked filled rubber shows significant time dependent stress relaxation as stress reduces significantly at the beginning of the holding time and then gradually levels off to a constant level with time. The unfilled natural rubber shows much less time dependence at the approximate same strain level as stress seems to change infinitesimally with time.

3. Material Modelling

3.1 Hyperelasticity

Denoting material co-ordinates by \( X \) and spatial co-ordinates by \( x(X) \), the deformation gradient \( F \) is defined by \( F = \frac{\partial x}{\partial X} \). Deformation of a material can be described by left Cauchy-Green deformation tensor \( B = F \cdot F^T \) or the right Cauchy-Green deformation tensor \( C = F^T \cdot F \).

Time independent large strain hyperelasticity is first considered. Here we assume that the long time (equilibrium) and the instantaneous response of rubber-like solids can be approximated as hyperelastic, thus having a unique stored strain energy density function \( W(B) \) that depends only on current strain. We shall therefore restrict attention here to isotropic materials. In this case, \( W \) may be expressed in terms of any three independent isotropic invariants of \( B \) defined by:

\[
I_1 = \text{tr}B \\
I_2 = \frac{1}{2} \left( [\text{tr}B]^2 - \text{tr}B^2 \right) \\
I_3 = \text{det}(B)
\]

(1)

Relation exists between Cauchy stress tensor \( \sigma \) and \( W \) can be expressed as [3]:

\[
\sigma = 2J^{-1} \frac{\partial W(B)}{\partial B} B ; \quad J = \text{det}(F)
\]

(2)

For incompressible, Isotropic material in isothermal condition, the relationship can be reduced to

\[
\sigma = -pI + \alpha_1 B + \alpha_2 B^2
\]

(3)

where the coefficients in this case are

\[
\alpha_1 = 2 \left( \frac{\partial W}{\partial I_1} + I_1 \frac{\partial W}{\partial I_2} \right) , \quad \alpha_2 = -2 \frac{\partial W}{\partial I_2}
\]

(4)
In the incompressible limit we have $J = I_3 = 1$, and the stress becomes indeterminate to within a hydrostatic stress $-\rho$ (to be found from boundary conditions).

In this work the strain energy function is assumed to be expressed by a polynomial series which can be written as:

$$W = \sum_{i,j=1}^{N} C_{ij} (I_1 - 3)^i (I_2 - 3)^j$$  \hspace{1cm} (5)

where $C_{ij}$ are parameters that can be determined from experimental data.

Under uniaxial loading, the relationship between the principal Cauchy stress and the stretch in loading direction $\dot{\lambda}_1$ under assumption of incompressibility can be written as:

$$\sigma_1 = 2\left(\dot{\lambda}_1^2 - \dot{\lambda}_1^{-2}\right) \frac{\partial W}{\partial I_1} + \left(\dot{\lambda}_1 - \dot{\lambda}_1^{-3}\right) \frac{\partial W}{\partial I_2}$$  \hspace{1cm} (6)

and the engineering stress in loading direction is

$$T_1 = \frac{\sigma_1}{\dot{\lambda}_1} = 2(1-\dot{\lambda}_1^{-3}) \left[ \dot{\lambda}_1 \frac{\partial W}{\partial I_1} + \frac{\partial W}{\partial I_2} \right]$$  \hspace{1cm} (7)

### 3.2 Viscoelasticity

For the small strain linear viscoelasticity, a thorough description of the subject can be found in many text books such as in the book by Ward [4]. For the finite strain, linear viscoelasticity, deviatoric part of Kirchhoff stress tensor at time $t$ denoted by $\mathbf{S}(t)$ can be expressed by [5]

$$\mathbf{S}(t) = \mathbf{S}^0(t) + \text{Sym} \left\{ \int_0^t \frac{d}{ds} \left( g_s^{-1}(s) \mathbf{F}(t-s) \mathbf{S}^0(t-s) \cdot \mathbf{F}(t-s) \right) ds \right\}$$  \hspace{1cm} (8)

$g_s$ is dimensionless relaxation function $g_s(t) = \frac{G_s(t)}{G_0}$, $G_s(t)$ is time dependent shear modulus and $G_0$ is instantaneous shear modulus. $s$ is dummy variable for $t$. $\mathbf{S}^0$ is instantaneous deviatoric Kirchhoff stress. $\mathbf{F}(t-s)$ is the deformation gradient of the state at $t-s$ relative to the state at $t$. Sym is symmetric operator to ensure that the stress remains symmetric. The volumetric behaviour is characterised by the incompressibility assumption.

The time dependent relaxation function can be defined using a Prony series expansion as

$$g_R(t) = 1 - \sum_{k=1}^{n} g_k \left( 1 - e^{-t/\tau_k} \right)$$  \hspace{1cm} (9)

where $\tau_k$ is termed the relaxation time. $g_k$ and $\tau_k$ are parameters to be determined from test data.

### 3.3 Visco-hyperelastic modelling

The hyperelastic and viscoelastic material model can be combined to yield a visco-hyperelastic material model. The instantaneous behaviour of rubber can be characterised by a hyperelastic model described through the elastic strain energy $W$.

The relationship in equation (7) was fitted to the up-loading uniaxial stress–strain data of carbon black filled rubber at crosshead speed of 300 mm/min shown in Figure 3, which we assumed that the stress-strain data can be approximated to the instantaneous response. It was found that by using the potential energy with polynomial series $N = 2$, very good fit to the experimental data can be obtained as shown in Figure 3. The corresponding parameters were obtained through a least square fitting method and are shown in Table 1.

![Engineering stress vs. Engineering strain graph](Figure 3)

In this work, the viscoelastic behaviour is assumed to be linear viscoelastic. The hyper-viscoelastic model was then implemented using the following keywords in the commercial FE code ABAQUS: 

- Hyperelasticity, $n=2$, moduli= instantaneous
- Viscoelastic, time=prony

The parameters describing viscoelasticity in equation (9) were found from using the visco-hyperelastic FE model to simulate the uniaxial stress relaxation experiment at strain level of 45% with the optimised hyperelasticity parameters and then the viscoelastic parameters were manually chosen until very good
fit to the data can be found. The stress relaxation data can be very well reproduced using Prony series with $n=3$ as shown in Figure 4. All the parameters for hyperelastic and viscoelastic were listed in Table (1).

The FE hyper-viscoelastic model with all the parameters in Table (1) was used to simulate uniaxial loading - unloading (cyclic) experiment at extension rate 300 mm/min using a plane stress element CPS4. The comparison of the simulations and experimental data with final strain levels of 26% and 62% are shown in Figure 5 and Figure 6 respectively. From these figures, the FE hyper-viscoelastic model can capture uploading curves very well but noticeably over predicts the unloading curves giving much less hysteresis loop than the experimental data. The discrepancies between the simulations and experimental data may arise because of the linear viscoelastic assumption used. The relaxation function in fact should not be only time dependent (linear viscoelasticity) but it should also be dependent of strain magnitude and strain history (nonlinear viscoelasticity). The nonlinear viscoelastic behaviour of carbon black filled natural rubber should be described instead if more realistic behaviour is to be obtained. The non-linear viscoelastic behaviour of particle filled natural rubber is very complex and is very much an active field of research. In the future, the authors aim to improve the hyper-viscoelastic model further to incorporate the nonlinear viscoelastic effect.

<table>
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<th>$C_{10}$ (MPa)</th>
<th>$C_{01}$ (MPa)</th>
<th>$C_{11}$ (MPa)</th>
<th>$C_{02}$ (MPa)</th>
<th>$C_{20}$ (MPa)</th>
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<td>4.708</td>
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Table 1: Parameters in the hyper-viscoelastic model

<table>
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<th>$g_1$</th>
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<th>$g_3$</th>
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<table>
<thead>
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<th>$\tau_2$ (s)</th>
<th>$\tau_3$ (s)</th>
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<td>100.0</td>
<td>1000.0</td>
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Figure 4. Simulation of the stress relaxatation behaviour using the viso-hyperelastic FE model.

Figure 5. Comparison of the visco-hyperelastic model prediction and stress-strain data obtained at 10th cycle of uniaxial tension cyclic loading up to 26% strain.

Figure 6. Comparison of the visco-hyperelastic model prediction and stress-strain data obtained at 10th cycle of uniaxial tension cyclic loading up to 62% strain.
4. Conclusions

A simple FE visco-hyperelastic model for describing finite strain uniaxial mechanical response of carbon black filled natural rubber was presented. The hyperelastic behaviour was represented by strain energy potential in the form of polynomial series. The finite strain viscoelastic behaviour was assumed to be linear viscoelastic and the relaxation function was represented by a Prony series. The model have some success in modelling the uniaxial tensile stress-strain curve during uploading and stress relaxation at a finite strain level. The model can depict the hysteresis response but in much lesser extent than that observed experimentally.

The constitutive modeling of the time-dependence of a particle filled elastomer is a complicated problem. The strain magnitude and history dependent relaxation function is needed if more realistic behaviour of carbon filled is to be depicted.

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Reference