Modelling the effects of various contents of fillers on the relaxation rate of filled rubbers

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Motivations

SBR with or without silica filler (Robisson, 2000)

Unfilled NR with or without crystallization (Bennani, 2006)

NBR with various carbon black contents (Omnes, 2007)

The higher the particles content, the higher the stress relaxation rate

What about ~100% fillers?
Motivations cont’d

✓ ECCMR 2007 (numerical homogenization)
  ✓ V. Jha et al. *Micro-structural finite element modelling of the stiffness of filled elastomers: the effect of filler number, shape and position*, pp. 165
  ✓ M. Naito et al. *3D modeling and simulation of micro to macroscopic deformation behavior of filled rubber*, pp. 127

✓ Applied to monotonous tensile loading (no viscous effect)

✓ Basic constitutive equations, with arbitrary materials coefficients
Content

- Background

- FE computations on 2D axi-symmetrical unit cell

- Extension to 3D: digitized microstructure

- FE computations on periodic cell

- Summary

- Future works
Small-strain linear viscoelasticity

Material coefficients

\[ K = 3000 \text{MPa} \quad G = 1 \text{MPa} \]
\[ \tau_1 = 0.01 \text{s} \quad g_1 = 0.3 \]
\[ \tau_2 = 0.2 \text{s} \quad g_2 = 0.1 \]

Loading step: imposed strain rate $0.2\text{s}^{-1}$

Stress-strain curve

Stress relaxation curve

Material coefficients

\[ K = 3000 \text{MPa} \quad G = 1 \text{MPa} \]
\[ \tau_1 = 0.01 \text{s} \quad g_1 = 0.3 \]
\[ \tau_2 = 0.2 \text{s} \quad g_2 = 0.1 \]
Small-strain linear viscoelasticity

Material coefficients

\[ K = 3000 \text{MPa} \quad G = 1 \text{MPa} \]

\[ \tau_1 = 0.01 \text{(s)} \quad g_1 = 0.3 \]

\[ \tau_2 = 0.2 \text{(s)} \quad g_2 = 0.1 \]

Loading step: imposed time 1s

Stress-strain curve

Stress relaxation curve

Loading step

K = 3000 MPa           G = 1 MPa
\[ \tau_1 = 0.01 \text{(s)} \quad g_1 = 0.3 \]
\[ \tau_2 = 0.2 \text{(s)} \quad g_2 = 0.1 \]

Material coefficients
Summary

✓ **Small strain viscoelasticity:**
  ✓ Controlled loading strain rate ➔ effect on relaxation rate
  ✓ Controlled loading time (actual) ➔ no effect on relaxation rate

✓ **Finite strain hyperviscoelasticity**
  ✓ Controlled loading time ➔ effect on relaxation rate (non linearity)
Unit cell 2D axisymmetrical simulation

✓ Meshes (axi-symmetrical)

\[ f = 10\% \]
\[ f = 20\% \]
\[ f = 40\% \]

✓ Particle radius: \( r = \frac{3f}{\sqrt{2}} \)

Inter-particle distance: \( h - 2r \)

✓ Loading

✓ Imposed time to loading: 1 second → 20% (uniaxial tensile)

✓ Constitutive relations

✓ Matrix: visco-hyperelasticity

✓ Particle: elasticity
Matrix: Finite-strain viscohyperelasticity – Inclusion: elasticity

Material coefficients

**Matrix**
Rivlin: $C_{10}=C_{01}=0.46\text{MPa}$, $C_{20}=0.15\text{MPa}$

$\tau_1 = 0.01\text{s}$, $g_1 = 0.3$

$\tau_2 = 0.2\text{s}$, $g_2 = 0.1$

**Inclusion**
$E=80000\text{MPa}$
$\nu=0.3$

Loading step to $\lambda=1.2$ : time 1s
Relaxation = 10s

Particles contents:
0%, 10%, 20%, 40%

Stress-strain curve

Stress relaxation curve

Loading history

Tire Expo Conference Köln 9-11/01/2010
Matrix: Finite-strain viscohyperelasticity – Inclusion: elasticity

Gage length effect: $l_0 \downarrow \Rightarrow \Delta l/l_0 \uparrow$
Investigation on the matrix under visco-hyperelasticity

**Material coefficients**

Rivlin: \( C_{10} = C_{01} = 0.46 \text{MPa} \quad C_{20} = 0.15 \text{MPa} \)

\( \tau_1 = 0.01 \text{(s)} \quad g_1 = 0.3 \)

\( \tau_2 = 0.2 \text{(s)} \quad g_2 = 0.1 \)

**Loading step: imposed time 1s**

Stress-strain curve

Stress relaxation curve

Material coefficients:

\( \tau_1 = 0.01 \text{(s)} \quad \tau_2 = 0.2 \text{(s)} \quad g_1 = 0.3 \quad g_2 = 0.1 \)
Results

✓ Experimental trend captured if:
  ✓ Matrix: visco-hyperelastic/ particle: elastic
  ✓ Imposed time to loading

✓ Gage length effect
  ✓ The higher the filler content, the smaller the inter-particle distance, the smaller the local initial gage length.

✓ Hyper-viscoelastic response
  ✓ The higher the particles content, the higher the stress level in the matrix, the higher the stress relaxation rate (due to non linear elasticity)
Extension to 3D: digitized microstructure (Numerical homogenization)

- Distribution of the inter-particle distance (dispersion)
- Effect of transverse inter-particle distance
- Cluster effects


- High amount of d.o.f.
- Parallel simulation under shear stress
- Time consuming
- Altering to periodic cell

Filler content: 20%

1600nm
Periodic cell simulation (effect of inter-particle distance)

✓ Meshes (tetrakaidecahedron)

✓ Loading

  ✓ Imposed time to loading: 1 second → 20%
  ✓ Boundary conditions at infinity (uniaxial tension or extension)

✓ Constitutive relations

  ✓ Matrix: visco-hyperelasticity
  ✓ Particle: elasticity

\[ f = \sqrt{3} \left( 2 + \frac{d}{R} \right)^{-3} \]
Matrix: Finite-strain viscohyperelasticity – Inclusion: elasticity

Material coefficients

Matrix
Rivlin: $C_{10} = C_{01} = 0.46\text{MPa}$, $C_{20} = 0.15\text{MPa}$

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Inclusion

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Particles contents:
0%, 10%, 20%, 40%

Stress relaxation curve

2D unit cell

3D Periodic cells

Tire Expo Conference Köln 9-11/01/2010
Matrix: Finite-strain viscohyperelasticity – Inclusion: elasticity

Average stresses in the matrix

\[ \sigma_{11} = \sigma_{22} = \sigma_{33} = 446\text{MPa} \]

Transverse displacement blocked at infinity!
Periodic uniaxial tension vs extension

- Small strain visco-elasticity
- Boundary conditions at infinity
  - No effect on the stress relaxation trend
  - Effect on the mean stress (triaxiality)
✓ Periodic cell vs unit cell, conclusion in agreement for
  ✓ Both global and local strain amplification (gage length effect)
  ✓ Stress relaxation rate
Outlooks

✓ **Meshing: inter-particle distance distribution**
   ✓ FE computation on digitized microstructure

✓ **Third phase (occluded gum, oriented amorphous interphase, crystallization)**
   ✓ Can be meshed on periodic cell
   ✓ Thickness/Consituitive equation of the interphase?

✓ **Constitutive models**
   ✓ Self consistent model (Omnes, 2007) Equivalent Homogenized Medium
   ✓ Statistical approach (Boukamel et al., ECCMR 2009, Dresden)

✓ **Asymptotic filler content**
   ✓ Saturation at 40%: electrical, mechanical percolation
   ✓ Short inter-particle distance → cavitation (tension), incompressibility (compression)…