Loss Amplification Effect in Multiphase Materials with Viscoelastic Interfaces

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Outlook

– Lamellar morphology systems
– Predicting effective viscoelastic responses of particulate morphology systems
  • Time domain finite element method
  • Four-phase composite sphere model
– High Impact Polystyrene (HIPS)
– Silica reinforced polymers
– Perspectives

\[
\gamma = \gamma_0 e^{i\omega t}
\]

\[
\mu = \mu' + i\mu''
\]

\[
\dot{E} = \mu''\gamma_0^2
\]
Lamellar Morphology

- Phase 1: $\mu_1 = b$
- Phase 2: $\mu_2 = a(1 + i\eta)$

Effective shear modulus

$$\frac{1}{\mu_{\text{eff}}} = \frac{f}{\mu_2} + \frac{1-f}{\mu_1}$$

- Asymptotic behavior for $a/b \to 0$
  - The maximum occurs at
    $$f = \sqrt{1+\eta^2} \left(\frac{a}{b}\right)$$
  - With a maximum value of
    $$\mu''_{\text{eff}} = b \left(\sqrt{1+\eta^2} - 1\right) / 2\eta$$

- Free & constrained layer damping treatments
  - widely used in practice

$b = 30 \text{ GPa}, a = 0.02 \text{ GPa}, \eta = 1$

\[
\frac{1}{C_{\text{eff}}} = \frac{f}{C_2} + \frac{1-f}{C_1}
\]

$C_1 = 80 \text{ GPa}$
$C_2 = 3.5(1 + i0.1) \text{ GPa}$
Morphology-Adaptive Meshes

Surface triangulation ↔ local curvature
Distance-based refinement

Volume mesh

Ill-shaped tetrahedrons:
Slivers, caps, needles & wedges

spacing between nodes ↔ local curvature
distance-based refinement

AAG, *Macromolecules* 2001, 34, 3081
Quality Refinement

- General purpose periodic mesh generator
  - sequential Boyer-Watson algorithm
  - $10^4$ nodes a second on a PC, independently of the mesh size
  - adaptive-precision floating-point arithmetic

Coating layers

Viscoelastic Responses

- Linear constitutive equation at a given frequency $\omega$

$$\sigma = D(\omega)(\varepsilon - \varepsilon_0) + D_d(\omega)(\dot{\varepsilon} - \dot{\varepsilon}_0)$$

- for systems with isotropic constituents ($K = \lambda + 2\mu/3$)

$$D = \begin{pmatrix}
\lambda + 2\mu & \lambda & \lambda & 0 & 0 & 0 \\
\lambda & \lambda + 2\mu & \lambda & 0 & 0 & 0 \\
\lambda & \lambda & \lambda + 2\mu & 0 & 0 & 0 \\
0 & 0 & 0 & \mu & 0 & 0 \\
0 & 0 & 0 & 0 & \mu & 0 \\
0 & 0 & 0 & 0 & 0 & \mu \\
\end{pmatrix}$$

$$D_d = \frac{\eta}{3} \begin{pmatrix}
4 & -2 & -2 & 0 & 0 & 0 \\
-2 & 4 & -2 & 0 & 0 & 0 \\
-2 & -2 & 4 & 0 & 0 & 0 \\
0 & 0 & 0 & 3 & 0 & 0 \\
0 & 0 & 0 & 0 & 3 & 0 \\
0 & 0 & 0 & 0 & 0 & 3 \\
\end{pmatrix}$$

- $\varepsilon_0$ is a prescribed function of time (e.g., a harmonic function)

- Principle of virtual work (weak form of the equilibrium equations)

$$\int_V \delta \varepsilon^T \sigma dV = 0$$
Steady-State Approach

- Resulting discrete equations for the nodal displacement vector $\mathbf{a}$
  
  $$ \mathbf{K}\mathbf{a} + \mathbf{C}\dot{\mathbf{a}} = \mathbf{f} $$

  - with $\mathbf{K} = \int \mathbf{B}^T \mathbf{D} \mathbf{B} \, dV$, $\mathbf{C} = \int \mathbf{B}^T \mathbf{D}_0 \mathbf{B} \, dV$, $\mathbf{f} = \int \mathbf{B}^T (\mathbf{D}\mathbf{\epsilon}_0 + \mathbf{D}_d \dot{\mathbf{\epsilon}}_0) \, dV$
  
  - and $\mathbf{B}$ standing for the strain-displacement matrix

- Steady-state solution
  
  $$ \mathbf{a} = (\mathbf{a}' + i\mathbf{a}'') e^{i\omega t} \quad \dot{\mathbf{a}} = (-\mathbf{a}'' + i\mathbf{a}') \omega e^{i\omega t} \quad \mathbf{f} = (\mathbf{f}' + i\mathbf{f}'') e^{i\omega t} $$

  - substituting and rearranging terms gives a saddle-point optimization problem

  $$ \begin{pmatrix} \mathbf{K} & -\omega \mathbf{C} \\ \omega \mathbf{C} & \mathbf{K} \end{pmatrix} \begin{pmatrix} \mathbf{a}' \\ \mathbf{a}'' \end{pmatrix} = \begin{pmatrix} \mathbf{f}' \\ \mathbf{f}'' \end{pmatrix} $$

  - Uzawa’s iterations, cg-solvers on normal equations, GMRES, etc.

  - but neither is suitable for large-scale ($10^6$ & larger) problems
Time-Domain Approach

- Imposing dynamic equilibrium at time $t_{n+1}$
  \[ K a^{(n+1)} + C \ddot{a}^{(n+1)} = f^{(n+1)} \]
  - and using the trapezoidal (“average velocity”) difference formula
    \[ \dot{a}^{(n+1)} = \frac{2}{\Delta t} (a^{(n+1)} - a^{(n)}) - \dot{a}^{(n)} \]
  - one obtains a Crank-Nicolson type, unconditionally stable algorithm
    - for stepping from $a^{(n)}$ at time $t_n$ to $a^{(n+1)}$ at $t_{n+\Delta t}$
      \[
        \left( C + \frac{\Delta t}{2} K \right) a^{(n+1)} = \frac{\Delta t}{2} f^{(n+1)} + g^{(n)}
      \]
      \[
        g^{(n)} = C \left( a^{(n)} + \frac{\Delta t}{2} \dot{a}^{(n)} \right)
      \]
    - at each time step, one should solve the above linear-equation system
    - iterative, preconditioned conjugate gradient solver
      - as both $K$ and $C$ are symmetric positive definite matrices
Extraction of Effective Properties

- Under external harmonic strain $\varepsilon = \varepsilon_0 \sin \omega t$
  - in the steady-state, the system stress is also harmonic $\sigma = \sigma_0 \sin(\omega t + \delta)$
  - the effective complex modulus $|\mu^*| = \sigma_0 / \varepsilon_0$
  - the effective loss factor $\tan \delta = \mu'' / \mu'$

$\mu_M = 0.01 + i 0.01$ [GPa]
$\mu_I = 1 + i 0.02$ [GPa]
$K_M = K_I = 3$ [GPa]

3 CPU days on a 1.6 GHz Itanium processor
Four-Phase Composite Sphere Models

- Pure dilation $\theta_0$ at infinity
  - Displacement vector $\mathbf{u} = (u_r, 0, 0)$
  - And is a function of $r$ alone
  - In spherical coordinates (in phase $i$)
    \[
    \text{div} \, \mathbf{u} = \frac{1}{r^2} \frac{d}{dr} \left( r^2 u_r^{(i)} \right) = \text{const} = 3a_i
    \]
  - Or $u_r^{(i)} = a_i r + \frac{b_i}{r^2}$
  - The radial stress $\sigma_{rr}^{(i)} = 3K_i a_i - \frac{4\mu_i}{r^3} b_i$

- Continuity at the boundaries
  \[
  \sigma_{rr}^{(i)}(R_i) = \sigma_{rr}^{(i+1)}(R_i) \quad u_r^{(i)}(R_i) = u_r^{(i+1)}(R_i)
  \]

- Self-consistent condition: the average dilation in the composite assembly is $\theta_0$
  - 6 equations, 6 unknown coefficients $\rightarrow K_{\text{eff}}$
Four-Phase Composite Sphere Models

- Pure shear \( \gamma_0 \) at infinity
  - Displacement vector \( u = (u_r, u_\theta, u_\phi) \)
  - In spherical coordinates (in phase \( i \))

\[
\begin{align*}
  u_r^{(i)} &= \left( A_i r - \frac{6v_i}{1-2v_i} B_i r^3 + 3 \frac{C_i}{r^4} + \frac{5 - 4v_i}{1-2v_i} D_i \right) \sin^2 \theta \cos 2\phi \\
  u_\theta^{(i)} &= \left( A_i r - \frac{7 - 4v_i}{1-2v_i} B_i r^3 - 2 \frac{C_i}{r^4} + 2 \frac{D_i}{r^2} \right) \sin \theta \cos \theta \cos 2\phi \\
  u_\phi^{(i)} &= -\left( A_i r - \frac{7 - 4v_i}{1-2v_i} B_i r^3 - 2 \frac{C_i}{r^4} + 2 \frac{D_i}{r^2} \right) \sin \theta \sin 2\phi
\end{align*}
\]

- Stress and displacement continuity at the boundaries
- **Self-consistent condition**: the average shear in the composite assembly is \( \gamma_0 \)
  - 12 second-order equations, 12 unknown coefficients \( \rightarrow \mu_{\text{eff}} \)

- Three-phase model: formulated by Kernel (1947), solved for \( \mu_{\text{eff}} \) by Christensen & Lo (1979)
  - Four-phase model was then solved by Herve and Zaoui (1991)
Spherical Interfaces

Bcc

Random

Composite Sphere Model

Effective medium

$R$ – inclusion radius
$\Delta$ – interface thickness

$R = R_2$
$\Delta = R_2 - R_1$
High Impact Polystyrene (HIPS)

\[
\begin{align*}
\mu_M &= \mu_I = 1 + i0 \text{ [GPa]} \\
\mu_L &= 0.01 (1 + i0.5) \text{ [GPa]} \quad (T_g) \\
K_M &= K_I = K_L = 3 \text{ [GPa]}
\end{align*}
\]
Spherical Interfaces

Shearing in the $xy$-plane

$$\mu_M = \mu_I = 1 + i 0 \text{ [GPa]}$$
$$\mu_L = 0.01 + i 0.005 \text{ [GPa]} \ (T_g)$$
$$K_M = K_I = K_L = 3 \text{ [GPa]}$$
Two Modes of Energy Dissipation

Energy-dissipation rate per unit volume \( \bar{E} \propto \langle \dot{\varepsilon}\sigma \rangle \)

Shearing in the xy-plane

\[
\begin{align*}
\varepsilon_0 &= \begin{pmatrix} 5 - i & 0 & 0 \\ 0 & -1.3 + i0.3 & 0 \\ 0 & 0 & -3.3 + i0.7 \end{pmatrix} \\
\varepsilon &= \varepsilon_0 e^{i\omega t} = \begin{pmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 0 \end{pmatrix} e^{i\omega t}
\end{align*}
\]

\( \Delta/R = 0.2 \)

\( \Delta/R = 0.007 \)
**Silica Reinforced Polymers**

<table>
<thead>
<tr>
<th></th>
<th>$K$ [GPa]</th>
<th>$\mu$ [GPa]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Silica</td>
<td>40</td>
<td>30</td>
</tr>
<tr>
<td>Solid polymer below $T_g$</td>
<td>4</td>
<td>$1 + i 0.02$</td>
</tr>
<tr>
<td>Polymer at $T_g$</td>
<td>$3.5 (1 + i 0.1)$</td>
<td>$0.01 (1 + i)$</td>
</tr>
<tr>
<td>Viscoelastic polymer above $T_g$</td>
<td>3</td>
<td>$0.001 (1 + i 0.1)$</td>
</tr>
</tbody>
</table>

![Graphs showing $\mu''$ and $\tan \delta$ vs. $\Delta/R$ for different materials.](image)
Lossy Syntactic Foams

- Epoxy matrix
- Rigid SiO$_2$ shell
- Lossy coating ($T_g$)
- Void
Perspectives

- Advanced materials for noise reduction and vibration damping
  - Epoxy matrixes filled with core-shell particles (syntactic foams)
    - such as glass micro-balloons coated with a lossy polymer layer
    - Short fiber and platelet reinforced polymers

- Numerical screening of the parameter space