Micromechanics-based Prediction of Thermoelastic Properties of High Energy Materials

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Objectives

- To predict thermoelastic properties of polymer bonded explosives at various strain rates and temperatures.
- To seek computationally efficient methods for the prediction of thermoelastic properties.
Outline

- Background
  - High Energy Materials
  - Micromechanics Methods
- Elastic Properties of Glass-Estane Mock Propellants
  - Bounds and Finite Element Estimates
  - Debonding
- Thermoelastic Properties of Polymer Bonded Explosives
  - Bounds and Analytical Approximations
  - Finite Element Estimates
  - Generalized Method of Cells Estimates
  - Recursive Cells Method Estimates
- Conclusions
High Energy Materials

- **Use:**
  - Propellants in Solid Rocket Motors
  - Explosives in Excavations
  - Detonators in Nuclear Devices

- **Examples:**
  - Ammonium Perchlorate and Aluminum Oxide
  - Polymer Bonded Explosives
Polymer Bonded Explosives

- Characteristics:
  - Particulate Composites
  - High Particle Volume Fraction (> 0.90)
  - Strong Modulus Contrast
  - Temperature and Strain Rate Dependence

- Examples:
  - PBX 9501: HMX\textsuperscript{1}, Estane 5703\textsuperscript{2} and BDNPA/F\textsuperscript{3}
  - PBX 9407: RDX\textsuperscript{4} and Exon-461
  - PBX 9502: TATB\textsuperscript{5} and KEL-F-800\textsuperscript{6}

\textsuperscript{1}High Melting Explosive
\textsuperscript{2}Segmented polyeurethene
\textsuperscript{3}Bis dinitropropylacetal/formal
\textsuperscript{4}Royal Demolition Explosive
\textsuperscript{5}Triaminotrinitrobenzene
\textsuperscript{6}Chlorotrifluoroethylene and vinylidene fluoride
PBX 9501

Microstructure of PBX 9501.

Components of PBX 9501.

<table>
<thead>
<tr>
<th>Material</th>
<th>Young’s Modulus (MPa)</th>
<th>Poisson’s Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>Particles (HMX)</td>
<td>17,700</td>
<td>0.21</td>
</tr>
<tr>
<td>Binder</td>
<td>0.7</td>
<td>0.49</td>
</tr>
</tbody>
</table>

Young’s Modulus of PBX 9501.
Micromechanics Methods

- Exact Results
  - Exact relation for coefficient of thermal expansion
  - Exact relations for effective elastic moduli

- Rigorous Bounds
  - Third-Order Bounds
  - Hashin-Shtrikman and Rosen-Hashin Bounds

- Analytical Approximations
  - Self-Consistent Scheme
  - Differential Effective Medium

- Numerical Approximations
  - Finite Elements
  - Generalized Method of Cells (semi-analytical)
  - Recursive Cell Method (renormalization-based)

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7 can be used to determine relative accuracy of numerical methods
8 provides bounds on the effective coefficient of thermal expansion
Elastic Moduli of Glass-Estane Mock Propellants
Glass-Estane Mock Propellants

- Why?
  - Not Explosive - Experiments Relatively Inexpensive
  - Large Range of Modulus Contrasts \( \frac{E_p}{E_b} \) - 8 to 10,000
  - Simple Geometry - Monodisperse Glass Beads in Binder
  - Low Filler Volume Fraction - 21% to 59%

- Approach
  - Two-Dimensional Finite Element Analysis
  - Three-Dimensional Moduli Determined From
  Two-dimensional Moduli

\[
\nu_{3D} = \frac{\nu_{2D}}{1 + \nu_{2D}} \\
E_{3D} = E_{2D}(1 - \nu_{3D}^2)
\]
Finite Element Estimates

- Discretization of Representative Volume Element (RVE)

- Application of Boundary Conditions

- Calculation of Effective Stiffness Matrix

\[
\langle \sigma_{ij} \rangle_V = C_{ijkl}^{\text{eff}} \langle \epsilon_{kl} \rangle_V
\]
Two-Dimensional Unit Cells

21% glass
- 5 Particles: 2.8x2.8 mm²
- 10 Particles: 4x4 mm²
- 20 Particles: 5.6x5.6 mm²
- 50 Particles: 8.9x8.9 mm²
- 100 Particles: 12.6x12.6 mm²

44% glass
- 5 Particles: 1.9x1.9 mm²
- 10 Particles: 2.7x2.7 mm²
- 20 Particles: 3.9x3.9 mm²
- 50 Particles: 6.1x6.1 mm²
- 100 Particles: 8.7x8.7 mm²

59% glass
- 5 Particles: 1.7x1.7 mm²
- 10 Particles: 2.4x2.4 mm²
- 20 Particles: 3.4x3.4 mm²
- 50 Particles: 5.3x5.3 mm²
- 100 Particles: 7.5x7.5 mm²
Effect of Unit Cell Size

Strain rate = 0.001/s and Temperature = 23°C.
Three-Dimensional Unit Cells
Are Two-Dimensional Unit Cells Adequate?

Three-dimensional Unit Cell and Slice
Two-Dimensional vs. Three-Dimensional

Similar Values of Young’s Modulus Obtained From Two- and Three-Dimensional Unit Cells

Two-dimensional vs. Three-dimensional Young’s Modulus at Strain Rate = 0.001/s
Bounds and Numerical Estimates

21% glass

59% glass
Debonds?

Unit Cell Containing 44% Particles by Volume - in Compression
Effect of Debonds

- Finite Element Estimates Higher Than Experimental Data - Even With Debonds
Conclusions

- Lower bounds are reasonable estimates of initial elastic moduli at low strain rates
- Two-dimensional finite element estimates are close to the lower bounds
- Considerable particle-binder debonding is required to match the predicted effective stiffness and the experimental data
Elastic Moduli of Polymer Bonded Explosives
### Bounds and Analytical Estimates for PBX 9501

#### Elastic Moduli and Thermal Expansion of Components of PBX 9501

<table>
<thead>
<tr>
<th>Material</th>
<th>Volume Fraction (%)</th>
<th>Bulk Modulus (MPa)</th>
<th>Shear Modulus (MPa)</th>
<th>Thermal Expansion (10^-5/K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>HMX</td>
<td>92</td>
<td>14300</td>
<td>5800</td>
<td>11.6</td>
</tr>
<tr>
<td>Binder</td>
<td>8</td>
<td>11.7</td>
<td>0.23</td>
<td>20</td>
</tr>
</tbody>
</table>

#### Bounds and Analytical Estimates of Properties of PBX 9501

<table>
<thead>
<tr>
<th></th>
<th>Bulk Modulus (MPa)</th>
<th>Shear Modulus (MPa)</th>
<th>Thermal Expansion (× 10^-5/K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PBX 9501</td>
<td>1111</td>
<td>370</td>
<td></td>
</tr>
<tr>
<td>Upper Bound</td>
<td>11306</td>
<td>4959</td>
<td>12.3</td>
</tr>
<tr>
<td>Lower Bound</td>
<td>224</td>
<td>68</td>
<td>11.6</td>
</tr>
<tr>
<td>Self-Consistent Scheme</td>
<td>11044</td>
<td>4700</td>
<td>12.9</td>
</tr>
<tr>
<td>Diff. Effective Medium</td>
<td>229</td>
<td>83</td>
<td>12.5</td>
</tr>
</tbody>
</table>
Elastic Moduli From Finite Element Analysis (FEM)
Validation of Approach

Two-Dimensional Finite Element vs. Differential Effective Medium
Validation of Approach

Two-Dimensional Finite Element vs. Three-Dimensional Finite Element

\( f_p = 0.7 \) \quad \( f_p = 0.75 \) \quad \( f_p = 0.8 \)
Validation of Approach

Two-Dimensional Finite Element vs. Three-Dimensional Finite Element

![Graph showing Young's Modulus vs. Particle Vol. Fractions for Two-Dimensional and Three-Dimensional Finite Element Methods.](image)

![Graph showing Poisson's Ratio vs. Particle Vol. Fractions for Two-Dimensional and Three-Dimensional Finite Element Methods.](image)
Models of PBX 9501

Manually Generated Microstructures

Around 89% particles meshed with triangles

Effective moduli of the six model PBX 9501 microstructures

<table>
<thead>
<tr>
<th></th>
<th>Expt.</th>
<th>Model RVE</th>
<th></th>
<th></th>
<th></th>
<th></th>
<th>Mean</th>
<th>Std. Dev.</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>1</td>
<td>2</td>
<td>3</td>
<td>4</td>
<td>5</td>
<td>6</td>
<td></td>
</tr>
<tr>
<td>E (MPa)</td>
<td>1013</td>
<td>116</td>
<td>126</td>
<td>130</td>
<td>42</td>
<td>183</td>
<td>192</td>
<td>132</td>
</tr>
<tr>
<td>ν</td>
<td>0.35</td>
<td>0.34</td>
<td>0.32</td>
<td>0.32</td>
<td>0.44</td>
<td>0.28</td>
<td>0.25</td>
<td>0.33</td>
</tr>
</tbody>
</table>
Models of PBX 9501

92% Particles By Volume

92% particles

E = 218 MPa  $\nu = 0.28$  E = 800 MPa  $\nu = 0.14$
Models of PBX 9501

Dry Blend of PBX 9501

Effective elastic moduli of the four models of the dry blend of PBX 9501

<table>
<thead>
<tr>
<th>Size (mm)</th>
<th>Young’s Modulus (MPa)</th>
<th>Poisson’s Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>FEM</td>
<td>Expt.</td>
</tr>
<tr>
<td></td>
<td>256×256</td>
<td>350×350</td>
</tr>
<tr>
<td>0.65</td>
<td>1959</td>
<td>968</td>
</tr>
<tr>
<td>0.94</td>
<td>2316</td>
<td>1488</td>
</tr>
<tr>
<td>1.13</td>
<td>2899</td>
<td>2004</td>
</tr>
<tr>
<td>1.33</td>
<td>4350</td>
<td>2845</td>
</tr>
</tbody>
</table>

0.65×0.65 mm²  0.94×0.94 mm²  1.13×1.13 mm²  1.33×1.33 mm²
Models of PBX 9501

Square Particles

Effective elastic moduli of microstructures with square particles.

<table>
<thead>
<tr>
<th>Size (mm)</th>
<th>Young’s Modulus (MPa)</th>
<th>Poisson’s Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>FEM (256x256)</td>
<td>Expt</td>
</tr>
<tr>
<td>3.6</td>
<td>9119</td>
<td>1013</td>
</tr>
<tr>
<td>5.3</td>
<td>9071</td>
<td>1013</td>
</tr>
<tr>
<td>9.0</td>
<td>9593</td>
<td>1013</td>
</tr>
</tbody>
</table>
Finite Element Estimates vs. PBX 9501 Experimental Data

- Red = Experiments
- Blue = FEM

Diagram showing the relationship between Young's Modulus (N/m²) and Strain Rate (1/s) with data points plotted for experiments and FEM simulations.
Conclusions

- Two-dimensional finite element models produces acceptable effective elastic properties

- Model geometry and mesh discretization plays a significant role in the predicted effective properties

- If a model is chosen in which the amount of stress bridging is optimum, excellent estimates of effective initial Young’s moduli can be obtained from finite element calculations
Elastic Moduli from the Generalized Method of Cells (GMC)
Generalized Method of Cells

- Why?
  - As accurate as FEM for fiber composites
  - More computationally efficient than FEM for fiber composites
Validation

\[
\begin{bmatrix}
\langle \sigma_{11} \rangle_V \\
\langle \sigma_{22} \rangle_V \\
\langle \tau_{12} \rangle_V
\end{bmatrix}
= \begin{bmatrix}
K_{\text{eff}} + \mu_{1}\text{eff} & K_{\text{eff}} - \mu_{1}\text{eff} & 0 \\
K_{\text{eff}} - \mu_{1}\text{eff} & K_{\text{eff}} + \mu_{1}\text{eff} & 0 \\
0 & 0 & \mu_{2}\text{eff}
\end{bmatrix}
\begin{bmatrix}
\langle \epsilon_{11} \rangle_V \\
\langle \epsilon_{22} \rangle_V \\
\langle \gamma_{12} \rangle_V
\end{bmatrix}
\]

\[
K_{\text{eff}} = 0.5(C_{11}^{\text{eff}} + C_{12}^{\text{eff}}) , \quad \mu_{1}\text{eff} = 0.5(C_{11}^{\text{eff}} - C_{12}^{\text{eff}}) , \quad \mu_{2}\text{eff} = C_{66}^{\text{eff}}.
\]

Comparison with effective moduli of square arrays of disks from Greengard and Helsing.
Models of PBX 9501

Manually Generated Microstructures

Model 1  Model 2  Model 3  Model 4  Model 5  Model 6

![Graph showing C_{11} (MPa) vs Model](image)
Two-Step GMC

GMC Discretization

First Homogenization Step

Homogenized RVE

Second Homogenization Step
Models of PBX 9501

Manually Generated Microstructures

Model 1  Model 2  Model 3  Model 4  Model 5  Model 6

Expt.
FEM
GMC – Grid Overlay
GMC – Two-Step
Models of PBX 9501

Dry Blend of PBX 9501

![Images of different dry blend models]

![Graph comparing experimental and modeled C_{11} values]
Stress Bridging

Effective properties of stress bridging models.

<table>
<thead>
<tr>
<th>Model</th>
<th>$C^\text{eff}_{11}$ (MPa)</th>
<th>$C^\text{eff}_{22}$ (MPa)</th>
<th>$C^\text{eff}_{12}$ (MPa)</th>
<th>$C^\text{eff}_{66}$ (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>FEM</td>
<td>GMC</td>
<td>FEM</td>
<td>GMC</td>
</tr>
<tr>
<td>Model A</td>
<td>16</td>
<td>16</td>
<td>16</td>
<td>16</td>
</tr>
<tr>
<td>Model B</td>
<td>336</td>
<td>19</td>
<td>343</td>
<td>19</td>
</tr>
<tr>
<td>Model C</td>
<td>4095</td>
<td>25</td>
<td>889</td>
<td>24</td>
</tr>
<tr>
<td>Model D</td>
<td>8992</td>
<td>8540</td>
<td>1361</td>
<td>32</td>
</tr>
<tr>
<td>Model E</td>
<td>10017</td>
<td>9042</td>
<td>10052</td>
<td>9042</td>
</tr>
</tbody>
</table>
Conclusions

- GMC is accurate for low particle volume fractions

- GMC is not recommended for high volume fraction composites such as PBX 9501
  - The predicted shear stiffness is too low
  - Unless a microstructure is chosen such that continuous particle paths exist across a subcell, the predicted normal stiffness is too low

- GMC is less computationally efficient than FEM for polymer bonded explosives because a large number of subcells is needed
Elastic Moduli from the Recursive Cell Method (RCM)
Recursive Cell Method

- Why?  
  ▶ Seek improvement over GMC for high volume fraction materials  
  ▶ More computationally efficient than FEM
Recursive Cell Method .. continued

- Similar to real-space renormalization methods
- Finite elements used to homogenize blocks of subcells

Boundary conditions used in RCM

\[ \nu_{\text{eff}}^{2D} = \frac{2C_{12}^{\text{eff}}}{(C_{11}^{\text{eff}} + C_{22}^{\text{eff}})} \]

\[ E_{\text{eff}}^{2D} = 0.5(C_{11}^{\text{eff}} + C_{22}^{\text{eff}})[1 - (\nu_{\text{eff}}^{2D})^2] \]

\[ \nu_{\text{eff}} = \nu_{\text{eff}}^{2D} / (1 + \nu_{\text{eff}}^{2D}) \]

\[ E_{\text{eff}} = E_{\text{eff}}^{2D} [1 - (\nu_{\text{eff}})^2] \]
Comparisons with Finite Element Estimates

Nine models of particulate composites

FEM vs. 2×2 RCM

FEM vs. 16×16 RCM
Models of PBX 9501
Effect of Increased Subcells Per Block

Model A

Model B

![Graph showing Young's Modulus vs. No. of Subcells/Block for different models](image-url)
An approximate estimate of the percolation threshold and the critical exponent can be obtained.
Summary and Conclusions

- RCM overestimates effective Young’s modulus and underestimates the Poisson’s ratio

- Increased subcells/block lead to improved estimates

- Better estimates for random microstructures

- RCM can be used to approximately identify percolation threshold in a computationally efficient manner
Overall Conclusions
Conclusions

- Thermal expansion can be predicted from bounds or analytical results

Elastic Properties:

- Bounds and analytical approximations are inaccurate at low strain rates and temperatures

- Two-dimensional finite element models provide reasonable estimates
  - Particle size distribution, mesh discretization and stress bridging affect prediction significantly
  - Debonding can also affect predicted values

- The generalized method of cells does not adequately account for stress bridging and is less computationally efficient than finite elements

- The recursive cell method accounts for stress bridging and is more computationally efficient than finite elements but overpredicts properties unless large blocks of subcells are renormalized
**Conclusion**

- Detailed simulation of actual microstructures using accurate numerical techniques is necessary to predict the effective properties of polymer bonded explosives.