Direct numerical prediction for the properties of complex microstructure materials

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Outlook

- Unstructured mesh approach
  - Short fiber composites
  - Clay/Polymer nanocomposites for barrier applications
  - Voltage breakdown in random composites

- Regular grid approach
  - 3D X-ray micro-tomography
  - Aluminum infiltrated graphite matrix composites
  - Pglass/LDPE hybrid materials
  - Cellular materials
Short fiber composites

- Polymers have a stiffness of 1-3 GPa
  - glass fibers 70 GPa
  - carbon fibers 400 GPa

- Short fiber reinforced polymers
  - fiber aspect ratio 10-40
  - volume loading 5-15%

- Can be processed by injection molding on the same equipment as pure polymers

Gear wheel

Acceleration pedal (Ford)
Local fiber orientation states

- Non-uniform fiber orientation states
  ⇒ non-uniform local material properties
    - stiffness
    - thermal expansion
    - heat conductivity, etc.

- Area with a high degree of orientation

- Area with a low degree of orientation
Direct finite element predictions

- Periodic Monte Carlo configurations
  - with non-overlapping spheres
  - with non-overlapping fibers


- Unstructured meshes (PALMYRA)
  - periodic morphology adaptive
  - $10^7$ tetrahedral elements
Validation

- Short glass-fiber-polypropylene granulate
  - Hoechst, Grade 2U02 (8 vol. % fibers)
  - injection molded circular dumbbells

- Image analysis
  - typical image frame (700x530 µm)

- Measured fiber orientation distribution
  - transversely isotropic
  - statistics of $1.5 \cdot 10^4$ fibers

- Measured phase properties
  - polypropylene matrix
    \[ E = 1.6 \text{ GPa}, \quad \nu = 0.34, \quad \alpha = 1.1 \cdot 10^{-4} \text{ K}^{-1} \]
  - glass fibers
    \[ E = 72 \text{ GPa}, \quad \nu = 0.2, \quad \alpha = 4.9 \cdot 10^{-6} \text{ K}^{-1} \]
  - average fiber aspect ratio $a = 37.3$
Validation

- Monte Carlo computer models
  - 150 non-overlapping fibers

- PJ Hine, HR Lusti, AAG

- AAG, PJ Hine, IM Ward
  *Comp. Sci. Tecn.* **2000**, 60, 535

- Fiber orientation distribution
  - compared to the measured one

- Effective properties
  
<table>
<thead>
<tr>
<th>Parameter</th>
<th>Numerical</th>
<th>Measured</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E_{11}$ [GPa]</td>
<td>5.14 ± 0.1</td>
<td>5.1 ± 0.25</td>
</tr>
<tr>
<td>$\alpha_{11}$ [$10^5 \cdot K^{-1}$]</td>
<td>3.1 ± 0.1</td>
<td>3.3 ± 1.5</td>
</tr>
<tr>
<td>$\alpha_{33}$ [$10^5 \cdot K^{-1}$]</td>
<td>11.7 ± 0.1</td>
<td>12.1 ± 0.2</td>
</tr>
</tbody>
</table>
Two step procedure

- Single fiber
  - unit vector $\mathbf{p} = (p_1, p_2, p_3)$

- System with $N$ fibers
  - 2nd order orientation tensor
    $$ a_{ij} = \langle p_i p_j \rangle $$
  - 4th order orientation tensor
    $$ a_{ijkl} = \langle p_i p_j p_k p_l \rangle $$

- Step 1: System with fully aligned fibers
  - numerical prediction for $C_{ijkl}$, $\alpha_{ik}$, $\epsilon_{ik}$, etc.

- Step 2: System with a given fiber orientation state
  - orientation averaging
  - quick arithmetic calculation
Orientation averaging

- Reference system with fully aligned fibers
  - transversely isotropic

- A system with given $a_{ij}$ and $a_{ijkl}$

- Effective elastic constants
  \[
  \langle \mathbf{C}_{\text{ref}} \rangle = \begin{pmatrix}
  C_{11} & C_{12} & C_{12} & 0 & 0 & 0 \\
  C_{12} & C_{22} & C_{23} & 0 & 0 & 0 \\
  C_{12} & C_{23} & C_{22} & 0 & 0 & 0 \\
  0 & 0 & 0 & C_{44} & 0 & 0 \\
  0 & 0 & 0 & 0 & C_{66} & 0 \\
  0 & 0 & 0 & 0 & 0 & C_{66}
  \end{pmatrix}
  \]

- Effective elastic constants
  \[
  \langle \mathbf{C} \rangle = B_1 a_{ijkl} + B_3 (a_{ik} \delta_{jl} + a_{il} \delta_{jk} + a_{jk} \delta_{il} + a_{jl} \delta_{ik}) \\
  B_2 (a_{ij} \delta_{kl} + a_{kl} \delta_{ij}) + B_4 \delta_{ij} \delta_{kl} + B_5 (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk})
  \]
  - $B_i$ are related to the coefficients of $\mathbf{C}_{\text{ref}}$

  - Direct & orientation averaging estimates
  - agree within 2-3%
Maximum entropy structures

- For transversely isotropic samples
- As the trace of $a_{ij}$ is 1, one needs only $a_{11}$
- Similarly, $a_{ijkl}$ is solely determined by
- The entropy of a system
  \[ S = -\sum_n p_n \log p_n \]
  $p_n$ is probability that the system is in state $n$
  \[ \theta_n \leq \theta \leq \theta_n + \Delta \theta_n \]
- Maximum of $S$ under a given $a_{11}$
- Comparison with image analysis data
Complex shape parts

- Steel molds (dies) are expensive
  - on the order of $20k and more

- Before any steel mold has been cut
  - mold filling flow simulations

- To optimize mold geometry & processing conditions
  - gate positions
  - flow fronts
  - local curing
  - mold temperatures
  - cycle times
  - etc.

- Software vendors: Moldflow, Sigmasoft, etc.
  - full 3D flow simulations instead of 2½ D
Computer-aided design of short fiber reinforced composite parts

- Method

- Short fibers:
  Hine, Lusti, AAG, *Comp. Sci.Tecn.* 2004, 64, 1081

- Spin-off company: MatSim GmbH, Zürich
  - Palmyra by MatSim, www.matsim.ch

- Acknowledgements
  - Professor U.W. Suter, ETH-Zürich
  - Dr. P.J. Hine, IRC in Polymer Science & Technology, University of Leeds
  - Dr. H.R. Lusti, ETH-Zürich
  - Professor I.M. Ward, University of Leeds
Voltage breakdown in random composites

- Dielectric parameters of pure polymers
  - dielectric constants
  - voltage breakdown
  - determined by chemical composition
  - and processing route

- Putting metal particles into polymers
  - increasing dielectric constants
  - decreasing voltage breakdown
    - local field magnifications

- Periodic unstructured meshes
  - morphology-adaptive

- Voltage breakdown mechanism
  - localized damage: pairs, triplets, etc
  - percolating path
Numerical procedure

- Numerical set up
  - matrix: $\varepsilon_M = 1$ & $E_c = 1$
  - inclusions: $\varepsilon_i = 10^6$

- Apply a very small $E$
  - such that all local fields $e$ are below $E_c$

- Gradually increase $E$
  - when somewhere in the matrix $e > E_c$
  - local voltage breakdown occurs
  - in the damaged sections, replace $\varepsilon_M$ by $\varepsilon_i$
  - go on

- Computer model with 27 spheres
  - here, sphere volume fraction $f = 0.3$

- Numerical predictions
  - overall dielectric constant: $\varepsilon_{eff} = 2.54$
  - breakdown field: $E_{eff} = 0.084$
Numerical predictions

- Numerical estimates
  - sphere volume fraction $f = 0.1$

- Composite voltage breakdown
  - arrestingly, ensemble minimum values representative already with only 8 spheres

- Overall dielectric constants
  - remarkably, uniform RVE size is very small
  - ensemble vs. spatial averaging

- Variable sphere volume loadings

<table>
<thead>
<tr>
<th>$f$</th>
<th>$\varepsilon_{\text{eff}}$</th>
<th>$E_{\text{eff}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1</td>
<td>1.37</td>
<td>0.12</td>
</tr>
<tr>
<td>0.3</td>
<td>2.57</td>
<td>0.03</td>
</tr>
<tr>
<td>0.5</td>
<td>5.12</td>
<td>0.008</td>
</tr>
</tbody>
</table>

- Technological aspects
  - relatively slow increase in $\varepsilon_{\text{eff}}$
  - rapid decrease in $E_{\text{eff}}$
Aluminium / Graphite Composites

- Squeeze casting process
  - graphite pre-form (porosity 14.5 vol-%)
  - infiltrated with aluminium melt (AlSi7Ba with 7 wt% Si & 0.25 wt% Ba)
- 3 phase composite: graphite (C), aluminium (Al), and pores
**3D X-ray tomography**

Swiss Light Source (SLS)

Beamlines at SLS

**XTM** (X-Ray Tomographic Microscopy)
- at Materials Science beamline of SLS
- beam energy of 10 keV
3D tomography microstructure

Pore

Al

C

200 x 600 x 600 pixels
pixel size = 0.7 µm

10 subvolumes:
25 x 25 x 25 pixel

10 subvolumes:
50 x 50 x 50 pixel

10 subvolumes:
100 x 100 x 100 pixel

9 subvolumes:
200 x 200 x 200 pixel
Effective conductivity

- FEM solution of Laplace’s equation
  \[ \text{div} \sigma(r) \text{grad} \phi = 0 \]
  - for nodal potentials
  - with position dependent \( \sigma(r) \)

- Technicalities:
  - serendipity family linear brick elements
  - iterative Krylov subspace solver
  - \( \sigma_{\text{eff}} \) from a linear response relation

- Implementation
  - GRIDDER by MatSim GmbH

25 x 25 x 25 pixel model
Representative Volume Element Size

Pglass / Polymer hybrids

- Pglass: 0.5 SnF$_2$ + 0.2 SnO + 0.3 P$_2$O$_5$
  - $T_g \approx 150$ C
  - can be melt processed at ca. 200 C

- 50/50 (by volume) Pglass/LDPE hybrid
  - melt processed at about 200 C
  - J. Otaigbe of Univ. of Southern Mississippi
  - measured stiffness:
    - LDPE 0.2 GPa
    - Pglass 30 GPa
    - composite 1.2 GPa

→ Why is the composite stiffness so low ?
→ Is the microstructure co-continuous ?

3D tomography microstructure

- Numerical predictions for $\sigma_{\text{eff}}$ assuming
  - either $\sigma_1 = 0$ and $\sigma_2 = 1$
  - or $\sigma_1 = 1$ and $\sigma_2 = 0$
- Both phases percolate
  $\Rightarrow$ co-continuous microstructure

400 x 275 x 200 pixels
pixel size $\sim 5 \, \mu m$
Property predictions (GRIDDER)

<table>
<thead>
<tr>
<th></th>
<th>LDPE</th>
<th>Pglass</th>
<th>Effective</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E$ [GPa]</td>
<td>0.2</td>
<td>30</td>
<td>7.1</td>
</tr>
<tr>
<td>$\alpha$ [$10^{-6}$/K]</td>
<td>150</td>
<td>12</td>
<td>58</td>
</tr>
</tbody>
</table>

- Predicted $E$ is 7 times larger than the measured one
- Hypothesis: disintegration of $P_{\text{glass}}$ phase
  - under the influence of residual thermal stresses
- Critical sections:
  - those with large von Mises stress & negative pressure
Residual thermal stresses

- Local stress tensor

\[
\sigma = \begin{pmatrix}
\sigma_{11} & \sigma_{12} & \sigma_{13} \\
\sigma_{12} & \sigma_{22} & \sigma_{23} \\
\sigma_{13} & \sigma_{23} & \sigma_{33}
\end{pmatrix}
\]

- Pressure

\[
p = \frac{1}{3} (\sigma_1 + \sigma_2 + \sigma_3)
\]

- Von Mises stress

\[
\tau = \sqrt{\frac{1}{2} \left[ (\sigma_1 - \sigma_2)^2 + (\sigma_2 - \sigma_3)^2 + (\sigma_3 - \sigma_1)^2 \right]}
\]
Stiffness of closed cell foams

- Real foams: \( \frac{E}{E_s} = C \left( \frac{\rho}{\rho_s} \right)^n \)
  - with \( 1 < n < 2 \)
- Kirchhoff plate theory
  - \( n = 1 \) reflects wall stretching
  - \( n = 3 \) reflects wall bending
- Sparse form solver
  - implemented in GRIDDER

\[ \rho = \text{CEE} \]
Materials with cellular microstructure

- Understanding structure – property relationships
  - both 3D X-ray tomography and model microstructures
  - mechanical, thermal, electrical, and other properties
Conclusions & Perspectives

- Unstructured mesh approach (PALMYRA)
  - Linear tetrahedral elements
  - Remarkably efficient for object based representations
  - Currently: spheres, spheroids, platelets, and spherocylinders
  - Locking problems with fluids and rubbers

- Regular grid approach (GRIDDER)
  - Linear brick elements
  - Very large grids, $10^9$ pixels and more
  - Appropriate for ordinary solids, rubbers and fluids
  - Property estimation companion for SCFT simulations

- For more on PALMYRA & GRIDDER technologies, visit www.matsim.ch